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A multidisciplinary perspective

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Learning from natural sediments to tackle microplastics challenges: A multidisciplinary perspective

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ABSTRACT

Although the study of microplastics in the aquatic environment incorporates a diversity of research fields, it is still in its infancy in many aspects while comparable topics have been studied in other disciplines for decades. In particular, extensive research in sedimentology can provide valuable insights to guide future microplastics research. To advance our understanding of the comparability of natural sediments with microplastics, we take an interdisciplinary look at the existing literature describing particle properties, transport processes, sampling techniques and ecotoxicology. Based on our analysis, we define seven research goals that are essential to improve our understanding of microplastics and can be tackled by learning from natural sediment research, and identify relevant tasks to achieve each goal. These goals address (1) the description of microplastic particles, (2) the

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Plastic pollution
Aquatic pollution
Sediment analogy
Microplastics

interaction of microplastics with environmental substances, (3) the vertical distribution of microplastics, (4) the erosion and deposition behaviour of microplastics, (5) the impact of biota on microplastic transport, (6) the sampling methods and (7) the microplastic toxicity. When describing microplastic particles, we should specifically draw from the knowledge of natural sediments, for example by using shape factors or applying methods for determining the principal dimensions of non-spherical particles. Sediment transport offers many fundamentals that are transferable to microplastic transport, and could be usefully applied. However, major knowledge gaps still exist in understanding the role of transport modes, the influence of biota on microplastic transport, and the importance and implementation of the dynamic behaviour of microplastics as a result of time-dependent changes in particle properties in numerical models. We give an overview of available sampling methods from sedimentology and discuss their suitability for microplastic sampling, which can be used for creating standardised guidelines for future application with microplastics. In order to comprehensively assess the ecotoxicology of microplastics, a distinction must be made between the effects of the polymers themselves, their physical form, the plastic-associated chemicals and the attached pollutants. This review highlights areas where we can rely on understanding and techniques from sediment research - and areas where we need new, microplastic-specific knowledge - and synthesizes recommendations to guide future, interdisciplinary microplastic research.

1. Introduction

Research on natural sediments has been conducted for decades (Wadell, 1933; Shields, 1936; Rouse, 1937) and can provide indispensable insights to guide our understanding of microplastics. Theoretical foundations and knowledge on natural sediments have guided research on the fate and transport of microplastics in the aquatic environment (Allen, 1985; Nizzetto et al., 2016; Horton and Dixon, 2017; Kooi et al., 2018), but most early studies in the field of ecotoxicology ignored direct comparisons between microplastics and natural sediments as they struggled to fully address the physical nature of the particles. Only recently, studies have started to compare the physical interaction of microplastics and sediments with organisms, focussing on e.g. the number of particles, surface area and particle shape (Schür et al., 2020; Scherer et al., 2020; Zimmermann et al., 2020). Moreover, microplastics research still has difficulties with fundamental tasks such as the description of particle properties (Rochman et al., 2019; Hartmann et al., 2019), transport (Hoellein et al., 2019; Petersen and Hubbard, 2021), or representative sampling (Cowger et al., 2021a; Liederhmann et al., 2018), each of which may benefit from concepts in sedimentary research. To delineate where concepts from natural sediments are or can be used for microplastics research and where we require further explorations, we review similarities and differences between natural sediment and microplastic particles regarding their particle properties, their transport behaviour, available sampling techniques and their ecotoxicology. This allows us to highlight knowledge gaps and identify future research questions that need to be addressed to significantly advance our still limited knowledge on microplastics.

2. Particle properties

While the description of sediment grains has a long history (Wadell, 1933; Sternberg, 1875; Wentworth, 1933; Krumbein, 1941), the most-suitable description of microplastic particle properties is still up for debate (Rochman et al., 2019; Hartmann et al., 2019; Kooi and Koelmans, 2019). This section highlights the similarities and the differences between the properties of microplastics and natural sediments regarding their size, shape and density, pointing out difficulties of microplastic particle description and indicating where knowledge from sedimentology can be useful for informing microplastics characterization.

2.1. Size

Determining the size of environmental particles is important to understand the fate of these particles in the field as well as correlating these data to exposure studies to gain understanding of the relative importance of size as a driver of bioavailability and toxicity for risk

assessment. Natural sediments are classified based on particle size ranges defining a 'class' (silt, sand, gravel) and grades specifying sub-ranges of a given class (e.g. fine sand, coarse sand). A commonly used particle size classification is the Udden-Wentworth grain size chart (Wentworth, 1933). Size definitions for microplastics are still debated (Hartmann et al., 2019), although the definition of particles with a diameter of less than 5 mm is often used (Arthur et al., 2008; Wagner et al., 2014; Koelmans et al., 2015), while other studies propose to use the micrometre scale (< 1 mm) to define microplastics (Browne et al., 2007; Andrady, 2015). The diameter hereby generally refers to the longest principal dimension of the particle, which is difficult to determine for microplastic particles due to their highly variable shapes (Kooi and Koelmans, 2019) and the currently used methods for size sorting, as described later on. The lower boundary for microplastics is debated even more, with values ranging from 0.1 µm up to 335 µm (Hartmann et al., 2019). However, 1 µm is often used to distinguish them from nanoplastics (Browne et al., 2007; Andrady, 2015; Gigault et al., 2018; GESAMP, 2016). If we compare the diameters of microplastics (here 0.001–5 mm) to the Udden-Wentworth scale, we end up with the corresponding classes of clay (< 0.004 mm), silt (0.004–0.063 mm), sand (0.063–2 mm) and gravel (2–63 mm).

The texture of natural sediments is described based on the volume or mass-based grain-size distribution (Tanner, 1995). The probability density function of the grain-size distributions can be either unimodal with one most frequently occurring grain size or polymodal showing multiple peaks in the grain-size distribution curve. The average grain-size of an unimodal distribution is described by the mode, median (d_{50}), and mean of the distribution curve (Boggs, 2009). Another key parameter to describe the texture of a sediment is sediment sorting. In a poorly-sorted sediment, grains exhibit a broader size range than in a well-sorted sediment, where a significantly narrower size range is seen (Boggs, 2009). Microplastic size distributions, on the other hand, have been described using count-based power law probability density functions, where a decreasing particle number concentration relates to an increasing particle diameter (Kooi and Koelmans, 2019; Cózar et al., 2014; Kooi et al., 2021). The slopes of these size distributions have been found to vary for different aquatic environments (Kooi et al., 2021). These differences are caused by different emissions as well as site and time specific hydrodynamics and processes, and are thus difficult to generalize (Kooi et al., 2021; Besseling et al., 2017; Yan et al., 2021). However, sediment-related values such as d_{50} and sorting have not been used to describe microplastics yet. Size distributions for microplastics can be used to align microplastic concentration values from different studies (Koelmans et al., 2020). Further efforts to apply particle size distribution descriptors and insights derived from the granulometry of natural sediments to that of microplastics will also have to contend with the differing unit bases of these systems (i.e. volume or mass-based vs. count based, respectively).

2.2. Shape

Although the shape of an object is a fundamental physical characteristic that influences its transport, it remains very difficult to characterize (Blott and Pye, 2007) - a problem that is well-known in both sediment and microplastic research. For natural sediment, shape descriptions are mostly based on geometric standards - such as ellipsoid, oblate spheroid (length \neq width = thickness), square/disc-shaped plates (length = width \neq thickness) and cylinder/rod (length \neq width = thickness) (Le Roux, 2005; Chamley, 1990). While shape characterization of natural sediments has been developed mostly to answer questions regarding transport properties (Dietrich, 1982), shape definitions in microplastic research are often used to identify the origin of the particles (Rosal, 2021). Primary microplastics - which are produced in sizes smaller than 5 mm - are mostly described as pellets and beads, while secondary microplastics - which fragment and degrade from bigger plastic items in the environment - include fragments, fibres, films and foams (Burns and Boxall, 2018). Although these shape categories are generally used in microplastic research to describe the particles, the actual particle shape in the categories can vary significantly (e.g. pellets may be cylindric, lenticular, or another shape) and some categories are not even shape-describing (e.g. foam). Accordingly, the microplastic community might have to rethink their shape categories to be able to clearly distinguish between different particle shapes, to increase the comparability of different studies and to incorporate shape into quantitative representations of transport processes. Cowger et al. (2020) proposed quantifying and reporting shape metrics for every individual particle to overcome this issue, while Metz et al. (2020) proposed to use the terms fragment and spherical/cylindrical pellet for particles with three dominant dimensions, the term foil for particle with two dominant dimensions and the term fibre for particles with just one dominant dimension. Kooi and Koelmans (2019) on the other hand proposed to use the relative ratio of particle sides (long:intermediate:short axes) to parameterize the different shape categories.

Another way to solve this issue is to use shape descriptors borrowed from sedimentology to define microplastic particles more accurately (van Melkebeke et al., 2020) and understand the role of the different shapes in the determination of particle size (Church, 2003). Here, the most important aspect of describing particle shapes lies in defining the principal dimensions (orthogonal long axis, L ; intermediate axis, I ; short axis, S), as the shape descriptors are mostly based on those dimensions (Blott and Pye, 2007). This includes, for example, the Wentworth flatness index (Wentworth, 1933) ($(L + I)/2S$), the Krumbein intercept sphericity (Krumbein, 1941) ($3I^{-2}SL^{-2}$), the Corey Shape factor (Corey, 1949) ($SL^{-1/2}I^{1/2}$), the Aschenbrenner working sphericity (Aschenbrenner, 1956) or the Janke shape factor (Janke, 1966). However, the process of defining and measuring the principal dimensions of irregular particles is highly error-prone, therefore Blott and Pye (2007) suggested for non-spherical sediments to follow the principle of the cube, where the principal dimensions are defined as the side lengths of the smallest imaginary cube which can contain the sediment grain. The principal dimensions also play different roles in the determination of characteristic particle size, depending on granulometric methodology. For natural sediments, sieving is most often used to measure the size of sand to gravel, and settling or optical techniques to measure the size of silt and clay. Sieve-based grain size is based on the retention of particles on the sieve, which is limited by I if all possible particle positions have been introduced to the sieve apertures (Tanner, 1995; Church, 2003), while settling and optical techniques can depend on both L and I (e.g. largest projected surface area), or all dimensions. As microplastics are more variable in their principal dimensions than most natural sediments (e.g. a fibre with $L > I = S$) and sieve-based size selection is a common approach for size definition, this aspect needs to be taken into account when defining microplastic particle dimensions. Settling, on the other hand, is not used to determine sizes of microplastics, since many plastics are buoyant, and the method would have to be adapted to this.

Regarding the description of the external geometrical form of natural sediments, Blott and Pye (2007) highlight four aspects that can - in combination - describe all kinds of sediment grains: form, roundness, irregularity and sphericity. Form describes the tridimensional characteristics and is defined by the ratio of the three principal particle dimensions (Sneed and Folk, 1958; Barrett, 1980). Roundness describes the degree of angularity (sharpness) of particle corners and edges (Powers, 1953). Irregularity is defined as the deviation of the particle shape from a regular body (Blott and Pye, 2007), and the sphericity is the degree to which a particle is similar to a sphere (Wadell, 1933). Only a combination of those descriptions is suitable to describe natural grains precisely, which seems to be the case for microplastics as well. For those, a combination of Corey shape factor, sphericity, circularity, elongation, flatness and aspect ratio is necessary to distinguish all microplastic shapes (van Melkebeke et al., 2020). Therefore, when describing microplastic particles, it is preferable to specify the principal dimensions, as a large number of shape descriptors can be identified on this basis, rather than relying on the conventional shape categories. However, we first need to find a way to reliably determine these principal particle dimensions.

2.3. Density

Sediment densities vary typically between 1.5 and 3 g/cm³, but, for simplicity, they are often assumed to be 2.65 g/m³ (Chamley, 1990), the density of quartz sand. Although coarse (e.g. sand, gravel) mineral particles are generally conveyed individually, fine, cohesive mineral sediments are often transported in association with other mineral and organic materials as aggregates or flocs (Droppo et al., 1997). Floc formation is dependent on the frequency and energy of particle interactions as driven by concentration and hydrodynamic conditions, the electrostatic charge of particle surfaces, and the concentration and character of dissolved ions, increasing with salinity (Winterwerp et al., 2006; Winterwerp, 1998), resulting in flocs that are larger than the size of their constituent mineral grains, but generally have lower densities (1.0 to 1.4 g/cm³) (Droppo, 2001; Faure et al., 2015; Maggi, 2005). The density of microplastics is mainly determined by the type of polymer that the particles consist of and varies between <0.05 g/cm³ for expanded polystyrene and 2.3 g/cm³ for polytetrafluoroethylene (Teflon) (Chubarenko et al., 2016). Microplastics can be transported in heterogeneous aggregates with other constituents such as organisms, suspended sediments, metal oxides and proteins as well (Yan et al., 2021), but the impact on the density seems to be more complex than for sediments. Heteroaggregation with inorganic particles such as suspended sediments and metal oxides results in a higher aggregate density (Wu et al., 2019; Leiser et al., 2020), while marine aggregation with other substances has been linked to a decrease in the aggregate density in comparison to the density of the individual microplastic particle (de Haan et al., 2019). While increasing salinity has been suggested to increase flocculation of microplastics (Andersen et al., 2021), the impact of the microplastic particle properties on floc formation has not been studied yet. Based on natural sediment floc formation and preliminary physical experiments, microplastic floc formation has been described by the fundamental metrics of collision frequency and the attachment efficiency (Besseling et al., 2017; Del Dommecq et al., 2021). The collision frequency is dependent on particle size, density and concentration and the attachment frequency depends on the particle surface characteristics and the water chemistry. Undoubtedly, flocculation plays a crucial role in the particle properties distributions of microplastics in the aquatic environment and needs to be studied in more detail (Wang et al., 2020; Alimi et al., 2018).

In addition to aggregation and flocculation, the density of microplastics can change over time due to fragmentation, degradation and biofouling, although these density changes are yet to be quantified (Skalska et al., 2020). Fragmentation and degradation ostensibly decrease the density of microplastics by creating additional pore volume

inside the particles (Ter Halle et al., 2016). The cracks that occur during fragmentation eventually lead to the particle breaking into smaller particles (Ter Halle et al., 2016), a process that has also been reported for natural sediments. Biofouling, the attachment of microorganisms and other organic particles on the surface of microplastics, can either increase or decrease the density of microplastic particles (Rummel et al., 2017). For buoyant microplastics, biofouling is suggested to increase density (Holmström, 1975; Ye and Andrady, 1991; Fazey and Ryan, 2016; Kaiser et al., 2017), while in the case of ‘high-density’ polyurethane microplastics (1.26 g/cm^3), biofouling was found to decrease particle density (Nguyen et al., 2020). This might be due to the density range of naturally occurring organic matter ($0.9\text{--}1.30 \text{ g/cm}^3$) (Harris, 2020). An increase in the density of initially buoyant plastic particles due to biofouling to more than 1 g/cm^3 (i.e. negatively buoyant) has been demonstrated in the aquatic environment after 6 weeks (Kaiser et al., 2017), 7 weeks (Ye and Andrady, 1991), or 3 to 4 months (Holmström, 1975). Compared with the residence time of plastic in the natural environment (decades to centuries), the time to form effective biofouling is very short. Given the significantly lower density of the biofilm, biofouling on the surface of high density sediment particles will decrease the overall density of bio-sediment (Shang et al., 2014; Fang et al., 2017). However, while microplastic fate can be strongly influenced by biofouling (particles that used to float start to sink), sediments will remain negatively buoyant.

3. Aquatic transport dynamics

In passing from fluvial to limnetic to the marine environment, sediments comprise different grain sizes from boulders and gravel upstream to sandy or clayey sediments closer to coastlines and oceans (Frings, 2008). While coarse grains are dominant in active areas with high flow velocities, fine and potentially cohesive sediments (i.e. mud and clay) can be found in hydrodynamically calm areas, e.g. close to the banks and on bar tops or within vegetation (Braat et al., 2017; van de Lageweg et al., 2018; Brückner et al., 2020; Browne et al., 2010; Zhang, 2017; Yao et al., 2019). The occurrence of microplastics has been linked to finer grain size fractions of sediments (Enders et al., 2019), suggesting that similar mechanisms govern their distribution. This empirical evidence raises questions on how the processes behind the transport, resuspension and deposition of different sediment sizes and microplastics relate, and if we can use our understanding of sediment transport processes to describe the transport of microplastics across aquatic systems. Although evidence exists for aeolian and terrestrial transport of microplastics (Dris et al., 2016), the bulk of the literature concerns sediment and microplastic transport by water, which is why we focus here solely on the latter. Moreover, we focus on transport by flow and currents, and neglect transport by wind waves for the most part.

3.1. Onset of motion

Several empirically derived sediment transport predictors can be used to describe resuspension of various types of sediments (Kleinhans, 2002). Generally, erosion can be estimated as a function of the hydrodynamic stress (i.e. excess shear stress) induced by currents or waves as well as a measure of the erodibility of the sediment often referred to as critical bed shear stress or critical velocity. Erosion occurs when the stresses induced by the hydrodynamic forces at the bed exceed the resuspension threshold of the sediment, which depends mainly on particle size, shape and density and can be estimated using the well-known Shields’ diagram (Shields, 1936; Coleman and Nikora, 2008; Dey et al., 2019). Although the Shields’ curve neglects interactions between multiple grain sizes both in suspension (e.g. flocculation) and at the bed (e.g. hiding, exposure, sediment sorting, cohesion), it is valuable for a first assessment of resuspension. While several field studies showed that microplastic abundance can be related to sediment grain size (Enders et al., 2019; Vianello et al., 2013; Ballent et al., 2016; Maes et al., 2018),

other research measured resuspension thresholds for microplastics, allowing us to describe them through similar metrics (Ballent et al., 2012; Waldschläger and Schüttrumpf, 2019a). The resuspension behaviour of microplastics is particularly dependent on the particle shape and size as well as the particle size distribution of the sediment in the bed. When comparing critical shear stresses of a variety of microplastic particles that were determined in physical model experiments with the Shields’ diagram (Shields, 1936), microplastics seem to be transported significantly earlier than calculated (Waldschläger and Schüttrumpf, 2019a). This indicates a higher mobility of microplastic particles compared to same-sized sediment grains, although this result depends on the used definition of on-set of motion (e.g. first particle movement, movement of 50% of grains) (Waldschläger and Schüttrumpf, 2019a). However, data on microplastic erosion dynamics is yet limited when compared to research on sediment dynamics from decades ago (e.g. van Rijn, 1984a). More in-depth studies, covering a wider range of microplastic parameters and testing different definitions of the onset of motion, are necessary to derive general resuspension behaviour expressions.

3.2. Transport modes of sediments and microplastics

Sediment and microplastic transport modes in the aquatic environment can be classified into surface, suspended, and bedload transport (Fig. 1) (Cowger et al., 2021a; van Rijn, 1984a; van Rijn, 1984b; Ancy, 2020). Surface load is the transport of larger, positively buoyant particles with high rising velocities that resist downward turbulent mixing, and are thus found primarily near the free surface. Suspended particulate transport is predominantly a phenomenon where the settling or rising of particles through the flow field is counteracted by the turbulence of the flow field itself. Bedload is the transport of larger particles with high settling velocities relative to turbulent fluctuations in the flow field that results in sliding, rolling and saltation of particles over the bed. Here we discuss the underlying mechanisms of these three transport modes for sediments and how they can be applied to microplastic transport.

3.2.1. Surface transport

Surface transport occurs when the particle buoyancy forces are greater than the turbulent mixing forces, a process that is rarely observed for natural sediments due to their high density. For microplastics on the other hand, surface transport has been observed in rivers (Lenaker et al., 2019) and is one of the most studied modes of transport of microplastics in the oceans (van Sebille et al., 2020). At large scales, surface currents can concentrate plastics in eddies in the centre of the oceans while windrows (Langmuir turbulence) have the same effect at smaller scales (van Sebille et al., 2020). Although such eddies can also exist in rivers, the concentration of plastics in them has not been studied in detail. Currently, sampling is often restricted to the water surface with the concentrations being extrapolated to the entire water column, with very limited knowledge about the vertical distribution of microplastics over the water column (Skalska et al., 2020). Buoyant microplastic concentrations seem to decrease with increasing water depth (Lenaker et al., 2019; Song et al., 2018; Gardon et al., 2021), thus to estimate the actual microplastics transport volumes in water bodies, it is essential to study the concentrations along the entire water column (Eo et al., 2019; Pabortsava and Lampitt, 2020).

3.2.2. Suspended transport

The governing equation describing suspended sediment transport is the advection-diffusion equation, which is typically derived using mass conservation in conjunction with Reynold’s decomposition and averaging, and Fick’s law (Fischer et al., 1979; Socolofsky and Jirka, 2005). The advection-diffusion equation for particle transport can be written as:

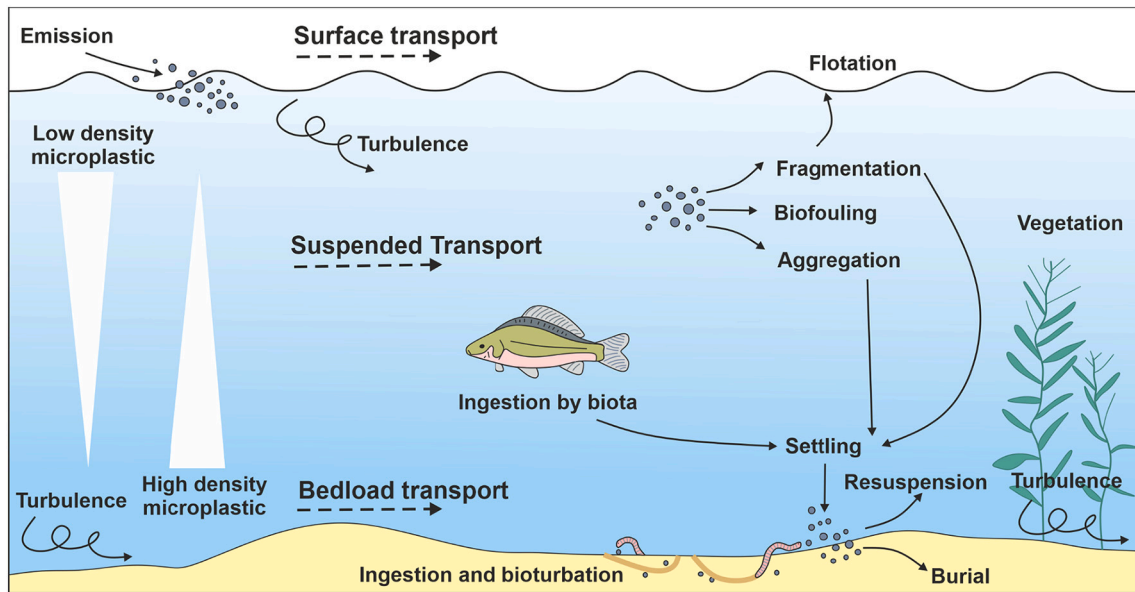


Fig. 1. Processes that act on and impact the transport of microplastics in aquatic systems. High- and low-density plastic is delineated at 1.0 g/cm^3 , width of white arrows represents qualitative microplastic density. Burial of sediment includes processes involving accumulation and migration. Modified from Skalska et al. (2020).

$$\frac{\partial \bar{c}}{\partial t} + \bar{u}_i \frac{\partial \bar{c}}{\partial x_i} = \frac{\partial}{\partial x_i} \left(D_i \frac{\partial \bar{c}}{\partial x_i} \right) + S \quad (1)$$

where \bar{c} is the particle concentration (e.g. microplastics), t is time, \bar{u} is mean velocity of the transported quantity, D_i is turbulent diffusivity, the overbar operator denotes time-averaging, x_i is the local coordinate in longitudinal (x), transverse (y) and bed-normal (z) directions, and S is a term for sources and sinks, which may comprise entrainment, deposition and infiltration processes of microplastics. Note that in Eq. (1), the turbulent diffusivity is a scalar, but it can also be a vector, for example if the diffusivity in the vertical direction is different from the horizontal directions, or even more generally the diffusivity can be a tensor (Spivakovskaya et al., 2007). It must be noted that turbulence is non-homogeneous and anisotropic in aquatic environments, which implies that turbulent diffusivities can vary in space and time. Eq. (1) assumes one-way coupling, meaning that particles are being passively transported by a turbulent flow without any feedback to the water flow.

Eq. (1) is a general three-dimensional advection-diffusion equation and can be solved numerically, given that flow velocities, turbulent diffusivities, sources and sinks are known. Simplified analytical solutions of Eq. (1) have been derived with relative success for both sediment and microplastic transport. For example, the assumption of uniform turbulence (Rouse, 1937; Dey, 2014) leads to:

$$D_i \frac{\partial \bar{c}}{\partial z} = -\bar{c}w \quad (2)$$

which can, based on the Boussinesq hypothesis and assuming the preservation of the logarithmic law of velocity distribution over the entire depth as described in Dey (2014), be integrated to:

$$\frac{\bar{c}}{\bar{c}_a} = \left(\frac{1 - \frac{z}{h} * \frac{a}{h}}{\frac{z}{h} * 1 - \frac{a}{h}} \right)^{\frac{Sc_T w}{\kappa u_*}} \quad (3)$$

where a is a reference level (length), h is the water depth (length), Sc_T is the turbulent Schmidt number (Gualtieri et al., 2017) (dimensionless), w is the settling/rising velocity (length/time), κ is the von Kármán constant (dimensionless, 0.41), u_* is the friction velocity (length/time). Note that in Eq. (3), turbulent diffusivities are expressed in terms of u_* , κ and the turbulent Schmidt number, which is the ratio between a

parabolic eddy viscosity and the turbulent diffusivity of microplastics, taking values between $Sc_T = 0.7$ to 1.4 , and a common value of $Sc_T = 1.0$ as a first guess in many environmental applications, see Gualtieri et al. (2017) and de Leeuw et al. (2020).

Eq. (3) is also known as the Rouse-equation and has been successfully applied to predict the vertical concentration of suspended microplastics (Cowger et al., 2021a) while a variety of other diffusion models exist (Dey, 2014). In rivers, the longitudinal distribution of suspended sediment and plastic particles is dominated by a process called longitudinal dispersion, characterised through combined effects of advection and vertical diffusion (Socolofsky and Jirka, 2005). Cook et al. (2020) showed that neutrally buoyant microplastic particles followed theoretical dispersion theory in laboratory uniform open channel flow, while it is known that longitudinal dispersion coefficients may vary significantly in natural river channels (Rutherford, 1994).

The settling and rising velocities are a key parameter in Eq. (3) and several studies have determined those for different microplastic particles in physical experiments (van Melkebeke et al., 2020; Khatmullina and Isachenko, 2017; Kaiser et al., 2019; Waldschläger and Schüttrumpf, 2019b; Waldschläger et al., 2020). A comparison with theoretical approaches from classical sediment transport for calculating settling velocities yielded only inaccurate results for microplastics (Waldschläger and Schüttrumpf, 2019b), whereas the use of shape-dependent drag models seems to be a better fit (van Melkebeke et al., 2020). This indicates that the shape of microplastics is possibly more significant for their transport than it is for natural sediments, also highlighted by the higher variation of aspect ratios (longest/shortest side length) of microplastics in comparison to natural sediments (Kooi et al., 2018; van Hateren et al., 2020). The importance of the particle shape can be explained by the significantly higher resistance depending on both preferential orientation (DiBenedetto et al., 2018), and more pronounced secondary movements in the sinking and rising process of the either rigid or deformable microplastic particles (van Melkebeke et al., 2020; Waldschläger and Schüttrumpf, 2019b; Waldschläger et al., 2020). Deformable particles have different settling characteristics from rigid particles as their shape changes during transport (Ardekani et al., 2017). This behaviour has already been shown for microplastic foils (Waldschläger et al., 2020), but a comparative description of the deformability of different microplastic particles has not yet been carried out.

Flocculation and aggregation can influence the settling behaviour of both sediments and microplastics. For natural sediments, the floc settling velocities are far smaller than for grains of the same size, but usually higher than that of the individual grains of which they are composed (Williams et al., 2008). Thus, flocculation tends to enhance deposition of fine sediments in aquatic systems. The same trend has been reported for flocculated microplastics (Alimi et al., 2018; Wang et al., 2021). Since microplastics have generally lower particle densities they can form relatively larger and denser flocs than the incorporated microplastics particles, which may result in higher deposition rates for given hydrodynamic conditions. As already discussed above, biofouling can either increase or decrease the density of microplastics, influencing the settling and rising behaviour in various ways. Kooi et al. (2017) hypothesise that microplastic particles exhibit oscillatory behaviour in the marine environment as a result of biofouling, in which the biofilm initially grows and causes the particle to sink. This effect diminishes towards deeper water layers due to the lack of sunlight, causing the particle to rise again. Additionally, size-specific removal of smaller microplastics due to biofouling from the surface has been observed in models (Kooi et al., 2017; Kvale et al., 2020a; Kreczak et al., 2021).

3.2.3. Bedload transport

Bedload transport denotes the near-bed movement of particles. The main modes of near-bed movement of microplastics are presumably the same as for natural sediments (Stubbins et al., 2021), namely sliding, rolling and saltation. Natural sediments transported as bedload (i.e. in continuous contact with the bed) differ mostly in diameter but are similar in density, and shape (mainly ellipsoidal due to abrasion and weathering). Bedload transport of microplastics, instead, needs a more detailed description in terms of particle characteristics due to their variety of shapes and densities, which leads to more complex transport (Kooi and Koelmans, 2019; Waldschläger and Schüttrumpf, 2019a). Although to date no study has intentionally focussed on bedload transport of microplastics, some studies theorized that bedload transport potentially generates smaller plastic particles through collisions with the bed (Morritt et al., 2014). Physical experiments adopted plastic grains rather than natural sediments to take advantage of their higher mobility starting from the '70s (Bettes, 1990), providing an extended body of literature that potentially holds some insights on microplastic near-bed transport. However, it must be noted that those experimental setups saw uniform microplastic particles moving on a microplastic bed, whereas in nature diverse microplastics move along a bed of natural sediments with significantly higher particle densities, presumably influencing transport. Thus, past research can specifically be used to elucidate the mechanics behind particle entrainment and near-bed motion of microplastics (Dey et al., 2019), but will not suffice to accurately parameterize bedload transport.

3.2.4. Turbidity currents

Another near-bed transport mode of importance for both microplastic and sediment transport are turbidity currents. Those currents have been recognized to distribute and bury large quantities of microplastics in seafloor sediments (Kane and Clare, 2019; Pohl et al., 2020). The loading of sediment from a river system into an open body of water could entrain and prograde natural and anthropogenic particles into the offshore as hyperpycnal (the flow in a river mouth that is denser than the water in the open water body receiving it and thus flows beneath the basin water) and hypopycnal (the flow in a river mouth that is less dense than the water in the open water body receiving it and thus flows above the basin water) flows (Warrick and Milliman, 2003; Renault and Gierlowski-Kordesch, 2010). In a series of experiments, Pohl et al. (2020) showed that a turbidity current carrying both microplastic particles and fibres developed a deposit relatively enriched in microfibrils implying that microfibrils were being trapped by settling sand grains at the base of the flow, while microplastic particles were transported on the bed. Notably, Pierdomenico et al. (2019) determined that a hyperpycnal

density flow can transport pollutants offshore into water depths of >1000 m. This suggests that submarine channels can act as long-term pathways of microplastics transport and deposition into the sea, extending from the canyon into the deeper basin and depositing beyond the channel mouth (Kane and Fildani, 2021). Microplastics initially transported by turbidity currents may be entrained by dense thermohaline bottom currents, which build large seafloor deposits known as contourites (Kane et al., 2020). Additionally, the microplastics-sediment transport mechanisms into lakes are poorly understood. This is concerning as density/turbidity currents in bedload dominated rivers play an important role in sediment, organic matter and nutrient dispersal within a lake (Marti et al., 2011).

3.2.5. Vertical transport in the sediment bed

Fine sediment and microplastic particles that are transported near the bed have the potential to be incorporated into coarser bed materials through hyporheic filtration (filtration into the transition area between river water and groundwater) (Karwan and Saiers, 2012). The migration of fine sediment particles into the hyporheic zone beneath the channel bed depends on horizontal and interstitial flows (Mathers and Wood, 2016), particle sinking properties (Schälchli and Schälchli, 1992), gravitational processes (Hauer et al., 2019) and water turbulence (Schälchli and Schälchli, 1992). While some knowledge exists on microplastic concentrations in hyporheic sediment (Frei et al., 2019; Drummond et al., 2020), only few studies have investigated depth-dependent concentrations in the sediment bed or hyporheic filtration of microplastics (Frei et al., 2019; Waldschläger and Schüttrumpf, 2020). They found that (i) vertical migration of microplastic particles into sediment beds occurs, (ii) smaller particle diameters lead to higher infiltration depths, and (iii) spherical particles migrate more easily than irregularly shaped microplastics. Infiltration depths depend on the mean grain size of the hyporheic sediment matrix and the ratio between microplastic diameter and mean grain size was identified as an indicator for the migration depth (Waldschläger and Schüttrumpf, 2020), which is similar to fine sediment hyporheic filtration, where the infiltration is dependent on the ratio between fine sediment diameter and bed sediment diameter (Schruff, 2018).

3.3. Modification of aquatic transport processes by biota

3.3.1. Impact of micro- and macrobenthic organisms on aquatic transport

Biota can both stabilize and disturb sediments. Benthic organisms can stabilize local sediments via the formation of biofilms or lead to the deposition of fine sediments in deeper soil layers via the formation of burrows by causing vertical mixing or ingestion (Herman et al., 2001; Widdows et al., 2004; Paarlberg et al., 2005; Monserrat et al., 2008). Biofilms enhance the sediment's stability against erosion by increasing the critical shear stress and can increase local cohesiveness, which affects the transport of the sediment (Brückner et al., 2020; Borsje et al., 2009). Similar to a stabilization of sediment by biofilms, microbenthic species likely stabilize microplastics in aquatic sediments as well (Hope et al., 2020), which affects their residence times and capacity to migrate into the deeper layers of the bed.

By contrast, macrobenthic organisms living on or in the sediment bed can disturb the sediment through bioturbation, for instance as a result of their feeding and mating behaviour (Coco et al., 2006; Le Hir et al., 2007; Cozzoli et al., 2018; Cozzoli et al., 2019). The destabilization of the sediment bed by bioturbation can result from the creation of burrows directly deteriorating the structure of the sediment matrix or indirectly through an increase of surface roughness by the creation of mounds or scrapes (Le Hir et al., 2007). Reworking of the sediment bed by bioturbation leads to increasing sediment fluxes towards the water column and subsequent dispersion or transport towards the ocean (Brückner et al., 2021). Although bioturbation can be described by a variation of the erodibility of the sediment (Cozzoli et al., 2019), uncertainties in quantifying the effects of macrobenthic species are related to the high

variability of species, their abundances, interactions and complex life-cycles (Le Hir et al., 2007). Vertical mixing into the sediment bed has been observed for conveyor-belt feeding behaviour of some deposit feeders (e.g. the worm *Tubifex tubifex*) through ingestion that leads to changes in distribution of sediment grains. While there is an increasing abundance of studies looking at the effects of ecotoxicology induced by microplastics, few studies discuss the change in microplastic resuspension, deposition and distribution in sediments mediated by macrobenthic organisms. Evidence exists however, for burial of microplastics into the deeper layers of the sediment through bioturbation and vertical mixing (Gebhardt and Forster, 2018; Coppock et al., 2021; Thit et al., 2020), suggesting that microplastic distribution in sediment can be affected by the reworking of sediments (Thit et al., 2020; Näkki et al., 2019) or by ingestion if sufficiently small.

More attention is required to quantify vertical transport of microplastics into the sediment bed, the interactions between benthic organisms and microplastic distribution and the role of biota in enhancing or reducing microplastic resuspension. The consequences of microplastic burrowing and distribution in sediments are crucial for predicting the transport and accumulation of these pollutants. Inevitably, the mediated resuspension of fines will naturally affect the resuspension of microplastics, which possibly results in increasing fluxes of microplastics and reduced residence times within the local sediment (Dürr et al., 2011).

3.3.2. Impact of vegetation on aquatic transport

The effects of vegetation on flow and sediment transport have been studied extensively on various spatial and temporal scales. While a breadth of papers discusses the effects of small-scale feedbacks between stems, local flow and sediment transport (Shan et al., 2020; Tinoco et al., 2020), an increasing number of studies investigate the emergence of scale-dependent feedbacks that determine transport rates and morphology from the patch to system-scale, such as the formation of tidal channels between vegetation patches (Temmerman et al., 2007) or the evolution of landforms (Mariotti, 2018; Gurnell et al., 2012; Nardin et al., 2018). Many of these studies showed that vegetation effects are highly species-specific, depending on emergent and submerged vegetation, flexibility of the stem, leaves, roots, and life-history traits (Baptist et al., 2007; Bij de Vaate et al., 2020).

In general, flow through a canopy can be described as a drag force that depends on the frontal area of the vegetation and vegetation-specific properties, such as stem flexibility or leaf area (Chapman et al., 2015). This force leads to a reduction in flow velocity, directly affecting the transport rates described by Eqs. (1) and (3). Vegetation also affects local turbulence and bed shear stresses, directly mediating resuspension and sediment transport rates (San Juan et al., 2019). At the same time, rooting structures enhance soil stability, increasing the resuspension threshold of the sediment (Abernethy and Rutherford, 2001). Around vegetation patches, flow velocities are accelerated, increasing resuspension and transport rates, thus creating near-field feedbacks between vegetation and flow with a spatial redistribution of particles and emerging morphological features (Brückner et al., 2020; Temmerman et al., 2007; Lera et al., 2019).

Despite the existing literature on vegetation effects on sediment transport, there are still many unknowns on the mediation of microplastic transport by vegetation (Zhang, 2017). Enhanced microplastics deposition is reported from both riverine and coastal ecosystems including riparian vegetation and freshwater wetlands (Yin et al., 2021; Ding et al., 2021), mangroves (Martin et al., 2020), salt marshes (Stead et al., 2020), seagrasses (Kreitsberg et al., 2021; Sanchez-Vidal et al., 2021; Huang et al., 2020), but also coral (Jeyasanta et al., 2020; Huang et al., 2021) and algae (Peller et al., 2021). Similar to sediments, species characteristics, abundance, shoot height, density, and leaf area index are considered to be key factors impacting the magnitude of flow alterations and microplastic accumulation and storage (Bouma et al., 2013; Helcoski et al., 2020). However, more studies are needed to investigate

vegetation-induced changes on (i) microplastics resuspension through altered turbulence, (ii) microplastic transport within and close to vegetation structures, and (iii) deposition rates both at the local and larger scale. Laboratory and field studies can elucidate if we can apply our understanding of sediment transport within canopies to microplastic transport, and can provide appropriate thresholds that allow for upscaling to the larger scale in numerical models.

3.4. Numerical transport modelling

Numerical models can be used to describe the spatial and temporal transport of a variety of particles in the aquatic environment (Papanicolaou et al., 2008), including microplastics and sediments, and add to our understanding of their distribution. Depending on the nature and the complexity of the problem that is addressed, the data availability for model calibration and verification, as well as the available resources (time & budget), models can be selected according to their spatial resolution: (Papanicolaou et al., 2008) zero-dimensional (0D, basin-output models), one-dimensional (1D, river/reach or water-column models), two-dimensional (2D, reach scale, floodplain, cross section-models) or three-dimensional (3D, including Lagrangian models). A variety of numerical models have been developed to model sediment transport, exchange processes such as settling and resuspension, and biotic effects, along with point and diffusive source pollutants attached to sediments (Papanicolaou et al., 2008; Peller et al., 2021). Likewise, numerical models have been developed to capture different microplastic transport processes (e.g. Nizzetto et al., 2016; Besseling et al., 2017; van Sebille et al., 2012). However, only few models on microplastic transport exist, meaning that the possibilities that numerical modelling offers - and that have already been exploited for sediment transport - are not yet exhausted. Table 1 compares sediment and microplastic modelling approaches and highlights the potential for microplastic research.

3.4.1. Zero-dimensions models

0D-models are time-dependent but have no spatial dependency. Basins are modelled as a box, where processes need to be parameterised. 0D-models are often implemented into higher-dimensional models to include basic processes (Cui et al., 2021). In sediment research, 0D-models have been used, for example, to predict soil loss from a field based on field parameters (Universal Soil Loss equation) (Stone and Hilborn, 2012), to model the concave longitudinal profile of a schematized alluvial river assuming a constant river width and length and a time-dependent bed composition (Franzoi and Nones, 2017), to investigate bathymetry changes in tidal environments over many decades with a simplified cohesive sediment transport model (Schoellhamer et al., 2008) or to model the aggregation of oil and natural sediments and to conduct sensitivity tests of fractal dimension and collision efficiency (Cui et al., 2021).

Several 0D-models have been built to estimate microplastic fluxes, including models that quantify plastic emissions from land to the oceans based on population density (Jambeck et al., 2015; Lebreton et al., 2017), models that calculate the probability for plastics to enter a river and subsequently the ocean based on geographically distributed data on plastic waste, land use, wind and precipitation (Meijer et al., 2021), or models that simulate fragmentation and settling on a whole-ocean scale (Jambeck et al., 2015; Koelmans et al., 2017).

The advantage of these simple relations is the low computational cost that allows for long term trend analyses in microplastic production, large spatial distributions and multiple scenarios. These models can be used as input for more complex models. Based on the already available 0D-models from sediment transport, modelling particle aggregation and flocculation of microplastics with natural sediments and biofouling seems to be an important and feasible step to improve our understanding of microplastic budgets in the future.

Table 1

Comparison of different approaches for numerical modelling of sediments and its potential for microplastics (MP) research.

Levels of approach	Fundamentals for sediment	Established achievements	Main limitations	Potential for plastics research	Exemplary references
0D-models (basin-output)	A basin can be modelled as a box. Processes need to be parameterised (calibration of many parameters) and output may depend strongly on the calibration procedure.	Useful for hydrological and water resources decision making	Bounded by limitations in parameterizations and their ranges for the studied basin processes.	MP budget quantification at the watershed scale	Stone and Hilborn, 2012
1D-models (river/reach)	Depth-averaged, commonly hydrostatic models. Vertical and transverse fluxes are neglected, and flow equations are solved commonly for the streamwise component. Models often require calibration (for instance, roughness or sediment transport rates).	Modelling of large river systems, representing unsteady flow effects and complex water systems. Represents a momentum-based modelling approach, yet a significant effort of model calibration may be needed to parametrize turbulence effects.	Subject to empirical relationships for sediment fluxes, hydrodynamics may not be captured in local areas where vertical fluxes and strong streamline curvature become important. Wetted cross-section considered on average terms.	Large-scale hydraulic transport of MP, but need for parametrisation of pickup, bedload transport and deposition.	Zech et al., 2008 ; Ferreira et al., 2009
1D-models (water column)	Water column models look at vertical transport only, neglecting horizontal changes.	Useful for studying settling rates and vertical concentration profiles. Water column models are lightweight, and can be run coupled to other models, such as flocculation.	Ignores horizontal processes.	Investigate timescales for settling, taking fouling and flocculation into account. Predict vertical concentration profiles.	Zhang et al., 2021a ; Dhamotharan et al., 1981 ; Gräwe, 2011
2D-models (e.g. reach scale, floodplains, cross section)	Depth averaged, allows better spatial discretization than 1D modelling across cross-sections (for instance, different velocities in floodplains). Vertical dimension is usually approximated while solving for horizontal fluxes.	Flood modelling in river and urban environments. Morphodynamic modelling of riverine and coastal areas achieved decades ago.	Similar to 1D-models but better representation of spatial variability (e.g. different velocities and depths across a cross-section)	Similar to 1D, but larger emphasis on flood plains and flooded areas (MP sinks). Implementation of multifractional microplastics. Distribution of microplastic transport into surface, suspended and bedload transport.	Hu et al., 2009 ; Soares-Frazão et al., 2012 ; Caldwell and Edmonds, 2014 ; Guerrero et al., 2015 ; Crosato and Saleh, 2011
3D ocean models (used on scales ranging from local (tens of kilometres) to global.	3D ocean models, such as ROMS and Nemo, are primarily designed to model the hydrodynamics of the ocean.	Some ocean models have modules for sediment modelling. Modelled ocean currents are also used as input to stand-alone models.	Relatively coarse resolution.	Can be used for Eulerian modelling of plastics transport, or provide modelled ocean currents for Lagrangian transport models.	Warner et al., 2008 ; Mountford and Morales Maqueda, 2019
3D / RANS models (reach/detailed structures-river interaction) relevant scales for scouring not solved, but presumed (turbulence modelling has a key role and main weakness)	Reynolds (time/ensemble) averaging. Flows are solved in <i>mean</i> terms, the effect of turbulence is parametrized through a turbulence model.	Accurate water-flow solution representing the mean flow dynamics. Some sediment transport modelling routines arising during the past decade.	The effect of turbulence is commonly approximated through an eddy viscosity, which is calculated based on (semi-)empirical turbulence models. This has an impact into the mean flow accuracy (minor) and into any turbulent process parametrised (major).	Turbulence can be modelled through general turbulence models (larger uncertainty in the wake of particles, for instance, and difficult to predict turbulent stresses).	Apsley and Stansby, 2008 ; Fuhrman et al., 2013
3D / LES models (very small scale - few meters), dunes formation, localized scouring, resolving relevant scales for scouring	Spatially filtered flow equations. Flow scales larger than the cell size are solved while a turbulent viscosity is responsible for sub-cell turbulence effects. Sub-cell turbulence is more isotropic, and more universal, which may lead to better suited turbulence modelling. Turbulence modelling dependence may vanish with reducing cell size.	Able to solve specific turbulence effects. Increasingly used with rising computational power.	Computationally more demanding than RANS. A LES is often required to solve at least 80% of the flow turbulence. Otherwise, it is considered an under resolved LES.	It can substitute or complement a bulk of hydraulic laboratory experiments. Reliable simulations with access to all hydrodynamic quantities	Zedler and Street, 2001 ; Nabi et al., 2013a ; Nabi et al., 2013b
3D / DNS models (interaction of a few particles with a predefined turbulent flow) allows real	Solves flow equations up to the smallest flow scales (i.e. Kolmogorov scale: 10^{-4} to 10^{-6} m in space 10^{-2} to 10^{-5} s in time). This is	Detailed studies on complex turbulence processes.	Computationally expensive, usually restricted to canonical problems with very small time and length extents.	Little gain compared to LES, yet considerably more computationally expensive. Primarily, useful to address fundamental matters.	Jain et al., 2021

(continued on next page)

Table 1 (continued)

Levels of approach	Fundamentals for sediment	Established achievements	Main limitations	Potential for plastics research	Exemplary references
insights into the interaction of particles with flow at scales comparable to the particle size and below	regarded as the most accurate level of flow modelling.				

3.4.2. One-dimensional models

Hydrodynamic 1D-models have been used for sediment transport since the early 1980s and are able to predict basic parameters (bulk velocity, water surface elevation, bed-elevation variation and sediment transport load and concentration) of a particular system (Papanicolaou et al., 2008). Application of 1D-models include simulations of flow and sediment transport along streams (Thomas and Prasuhn, 1977; Karim and Kennedy, 1982; Chang, 1984; Schick et al., 1998), mobile-bed dynamics (Holly et al., 1990), predictions of grain size distributions and bedload rates in rivers (Papanicolaou et al., 2008) and vertical distribution of suspended sediment and oil-sediment aggregates in the marine and fluvial environment (Cui et al., 2021). For a detailed description of the numerics of particle-based water-column models, see for example Gräwe (2011) and Nordam et al. (2019).

1D microplastic transport models have been developed for river (sub-)catchments, assuming homogeneous mixing of plastics in the water column (Nizzetto et al., 2016; Besseling et al., 2017; Unice et al., 2019). These models include processes like aggregation, degradation, settling and resuspension, and can be used for hotspot analysis, quantification of catchment emissions and a better understanding of the relative importance of different processes. However, as parameterization of most of the mentioned processes has not been derived for microplastics yet, caution is required with regard to the validity of the models' assumptions and results (Wichmann et al., 2019). Other 1D models focus on the vertical distribution of microplastics (Cowger et al., 2021a), the effect of biofouling on the settling of particles (Kooi et al., 2017), or the effect of wind-mixing on the vertical distribution of particles in the upper part of the ocean water column (Kukulka et al., 2012). Understanding the vertical distribution of microplastics is relevant both in terms of predicting the mean transport rates across a profile along a river, but also to estimate particle concentrations to better describe particle interactions, diffusion into the bed and ecotoxicological consequences.

The big advantage of 1D-models is the low computational requirements in comparison to 2D and 3D-models as well as their simplicity of use (Papanicolaou et al., 2008), while being more process-oriented than 0D-models. They are useful to understand transport rates and concentrations along the vertical or horizontal profile, describing in more detail where sediments and microplastics are transported (Rouse, 1937; Nizzetto et al., 2016; Cowger et al., 2021a; Kooi et al., 2017). Major assumptions of 1D-models are the absence of lateral and vertical exchange of particles across the channel and floodplains, or the assumption of horizontal homogeneity in the case of water-column models. Based on the already available 1D-models from sediment transport, bedload microplastic transport could be addressed in the future, but it would require specific empirical relationships.

3.4.3. Two-dimensional models

Since the early 1990s, more 2D sediment transport models have been used, as they allowed easy data input and visualization of the modelling results (Papanicolaou et al., 2008). As opposed to 0D- and 1D-models, 2D approaches allow for a more detailed representation of the lateral transport field. The vertical component is still considered in average terms and thus these models are often referred to as 2D depth-averaged models. Applications of 2D-models include investigations of the transport of radionuclides (Walters et al., 1982; Voitsekhoveritch et al., 1994), simulations of sediment transport processes and bed-level changes in

limnic, fluvial and coastal environments (Papanicolaou et al., 2008; Walstra et al., 1998; Niyayati and Maraghei, 2002; Czuba et al., 2015), predictions of sediment transport rates (Danish Hydraulic Institute, 1993) and investigation of the effects of local changes of the waterflow, e.g. by woody debris, rocks and anthropogenic structures, on sediment transport (Danish Hydraulic Institute, 1993; Wu et al., 2005; Jia and Wang, 1999). They can capture cross-sectional variations in morphology and velocity and can therefore estimate variations in transport rates across different water depths and lateral exchanges (Papanicolaou et al., 2008). Furthermore, 2D-models can predict total sediment transport load as well as multifractional sediment transport and can decompose the total sediment load into bedload and suspended load (e.g. Jia and Wang, 1999; Spasojevic and Holly, 1990; Lee et al., 1997; Chang, 1998).

2D mass balance models that predict microplastics concentrations and emissions on a catchment scale have been made both for Europe (Siegfried et al., 2017) and the world (van Wijnen et al., 2019). These steady-state models consider removal via settling and/or degradation, and in the case of van Wijnen et al. (2019) also fragmentation. Several 2D-models have been made that describe microplastic concentrations and processes in lakes (Cable et al., 2017; Hoffman and Hittinger, 2017; Mason et al., 2020). Some of these models include beaching as a removal process (Hoffman and Hittinger, 2017; Mason et al., 2020). A few of the early models of ocean plastic transport were 2D Lagrangian particle models (van Sebille et al., 2012; van Sebille et al., 2015; Maximenko et al., 2012). These models used surface tracers to predict how plastics move across the surface of the ocean. River networks have also been models regarding microplastic concentration introduced by point sources (Schmidt et al., 2020).

Based on the already available 2D-models from sediment transport, possibilities arise to model multifractional microplastic transport to include the highly varying particle properties of microplastics (size, shape, density) and observe their impact on microplastic transport. However, to implement those varying particle properties and their impact on the transport, we first need to parameterise these variables and processes. Additionally, the distribution of the total microplastic transport into surface, suspended and bedload-transport could be investigated in the future. This could be based on the probability function for microplastic density, diameter and shape developed from Kooi and Koelmans (2019) and Kooi et al. (2021). Differently from mineral sediment transport, microplastic particles may experience physical changes at the transport scale represented by these models.

3.4.4. Three-dimensional models

When looking at complex flow situations, for example around hydraulic structures, only 3D-models are able to represent the physics and to predict sediment transport (Papanicolaou et al., 2008) with adequate accuracy. Applications for sedimentary 3D-models include simulations of flow and sediment transport processes (HydroQual, 1998; Hamrick, 1992; Admass, 2005), simulations of sedimentation on bends, crossings and distributaries (Spasojevic and Holly, 1994), simulations of water quality (Jacobsen and Rasmussen, 1997; Delft Hydraulics, 1999) and simulation of the transport of chemicals (Bierman et al., 1992; Gu and Chung, 2003). Depending on the used model, simulations of movable riverbeds, bedforms, sediment sorting and armouring processes (Olsen, 1994), flocculation models for cohesive sediment (Winterwerp, 1998) as well as exchange of metals between the bed sediment and the water

column (Danish Hydraulic Institute, 1993) and implementation of parameters such as salinity, temperature, suspended sediment and their influence on density (King, 1998) are possible. These simulations can range from local to global scales (Papanicolaou et al., 2008). Similar to 2D-models, some 3D-models are able to predict sediment transport of sediment mixtures (Hamrick, 1992; Spasojevic and Holly, 1994; Olsen, 1994; Song and Haidvogel, 1994).

Both Eulerian and Lagrangian approaches can be used in 3D-models. While Eulerian modelling characterises how a particle concentration changes at a fixed grid of points, or cells, the Lagrangian description, also known as ‘particle tracking’, simulates particle transport by modelling the position of a collection of (numerical) particles. Mathematically, Eulerian and Lagrangian models are equivalent, and which formulation is most suitable will depend on the scenario to be modelled. Initially, Lagrangian drifters have only been used to model the surface transport of passive plastics particles in the oceans (van Sebille et al., 2012; Maximenko et al., 2012). Since then, Lagrangian models approximated biological transport by spatially constant removal rates and modelled non-conservative particles (Isobe et al., 2019), subsurface advection (Wichmann et al., 2019), as well as implemented a 1D-biofouling model (Kooi et al., 2017) to estimate sinking timescales for biofouled, initially buoyant microplastics in the global oceans (Daily and Hoffman, 2020). Daily and Hoffman (2020) also modelled the distribution of microplastics in the water column and the sediment of Lake Erie by using a Lagrangian transport model that implemented advection, density-driven sinking, and turbulent mixing. 3D Lagrangian methods can also be applied to smaller scales, and have been used to study the fate of non-buoyant microplastics in the surf-zone of beaches (Jongedijk et al., 2020). Eulerian models have so far been used to assess the effect of buoyancy and idealized removal on the global microplastic transport by implementing particles with different densities (Mountford and Morales Maqueda, 2019), as well as to embed microplastics in an earth system model that includes coastlines and major shipping lanes as input sources and biological interaction - aggregation of microplastics with marine snow and ingestion of microplastics by zooplankton - as a possible sink (Kvale et al., 2020b). Mountford and Morales Maqueda (2019) have thereby implemented the 0D-model by Jambeck et al. (2015) and van Sebille et al. (2015) on plastic emissions into their model, highlighting the possibilities to further develop low-dimensional models.

Several processes that might impact microplastic transport are not implemented in 3D-models yet. Wichmann et al. (2019) identify a number of processes that are not implemented in their model, but which presumably have an important influence on the transport behaviour of microplastics: particle properties such as composition, size and shape of microplastics, time-dependent processes such as biofouling, fragmentation and degradation, beaching of microplastics on coasts, e.g. by breaking waves, as well as oceanographic phenomena such as upwelling and tides. However, the temporal and spatial influences of most of these processes on microplastics transport have not yet been parameterized, so that an implementation in numerical models is accompanied by large uncertainties (Wichmann et al., 2019). Mountford and Morales Maqueda (2019) point out the importance of implementing microplastic-sediment interactions to improve their model in the future. Lobelle et al. (2021) highlights the need to implement particles with different shapes and fragmentation properties as soon as there are mathematical descriptions for them. Based on the already available 3D sediment models, the implementation of multifractional microplastics should be possible in the near future to improve the transport modelling of diverse microplastics. Additionally, the interaction between turbulence in different flows on different microplastic particles could be investigated by high-fidelity 3D-models (Large Eddy Simulation (LES) and Direct Numerical Simulation (DNS), see Table 1).

4. Environmental sampling techniques

Initial studies that reported microplastics in the environment did so

incidentally while sampling for other substances, thus protocols and observations are shaped by this historic legacy (Jongedijk et al., 2020). However, to understand the fate of microplastics within the environment, it is essential to collect representative environmental samples. Furthermore, ground-truthed environmental concentration values are necessary to validate models displaying microplastic emissions, transport pathways, and hotspots (van Emmerik et al., 2019; Weiss et al., 2021). Moreover, microplastics can be used in terms of geochronology. The detection in a sediment layer offers information on the maximum deposition time. Since the mass production of plastics started in the 1950s, prior deposition in the environment is very unlikely (Weber and Lechthaler, 2021). In addition, using contaminants in sediments as tracer to reconstruct floodplain chronology is a common technique regarding heavy metals and was transferred to microplastics where similar patterns in floodplain sediment deposition could have been shown (Lechthaler et al., 2021).

Since there are already numerous techniques for sampling the water column and sediment, an evaluation of their transferability is a new and important research aspect. Here, the suitability of these techniques for microplastic sampling, as well as detection and classification (shape, colour, size, polymer) of microplastics in sediment deposits, the water column, snow and ice are discussed, resulting in the identification of areas to be addressed in future research.

4.1. Field sample collection

The choice of field sampling technique depends on the environmental matrix and the portion of the microplastics size distribution targeted by the sampling plan. A variety of techniques have traditionally been deployed to sample sediment deposits with grain sizes comparable to those of microplastics (clay, silt, sand and gravel) (Table 2). Bulk sediment samples, collected through grab (e.g. Ponar or Van Veen grab), coring, trapping (Storlazzi et al., 2011; Tidjani et al., 2011), or drilling techniques (Kondolf et al., 2003), can equally be used to assess natural sediment or microplastics. The differentiation of target material is later achieved in the laboratory during separation. Similar to sediments, snow and ice sampling for microplastics can be conducted much as with sediment sampling through grab, core or drill sampling, and faces many of the same benefits and challenges for each technique (Bergmann et al., 2019; La Kanhai et al., 2020; Kelly et al., 2020). In aquatic environments, differences in techniques may be necessary when samples are being sourced at different water depths. Suspended sediment samples are typically collected isokinetically, where the suspended sediment mixture undergoes no change in speed or direction as it enters the mouth of the sampler, either integrated across channel depth or depth and width (i.e. flow integrated), or at a given depth using horizontal water samplers such as the Van Dorn (Wren et al., 2000; Davis, n.d.). Suspended sediment samples for microplastic analysis have been also collected using sediment traps (Lorenz et al., 2019; Bagheri et al., 2020; Fraser et al., 2020; Rios Mendoza et al., 2021). Microplastics on the other hand are sampled using filtration (i.e. using nets, sieves or filters), or are collected as grab sample (i.e. using a bucket). Microplastics concentration and character can vary widely for all environmental media (e.g. $<10^{-3}$ to $>10^5$ particles per litre), resulting in representative sample sizes that range from less than 1 l to multiple m^3 , depending also on the targeted microplastics and scientific questions (Shahul Hamid et al., 2018).

4.1.1. Possible routes for advances in sampling using known sediment techniques

Multiple approaches to sediment sampling and characterization including in-situ and remote optical techniques have been used to monitor fluvial and marine sediment transport for decades (Lynch et al., 1994), with notable recent advances (Czuba et al., 2015). Optical turbidity sensors have been used since the mid-20th century for continuous, point-based monitoring of suspended sediment

Table 2

Comparison of field sample techniques for sediments transferred to microplastics (MP).

Matrix	Environmental settings	Sampling technique	^a Contamination sources	^b Particle size range	^d Suitability for MP sampling	Exemplary References
Sediment deposit	terrestrial, aquatic	drill, core, trench	plastic core/casing liners, components	all	high, analysis of different layers possible without cross-contamination	La Khanh et al., 2019 (core), Lechthaler et al., 2021 (trench)
		surface grab		all, potential fine MP loss via dewatering	moderate, accumulation rate error from inconsistent sample geometry	Lorenz et al., 2019
Water column	aquatic	net	net, carry-over from previous samples	> mesh size: generally $\geq 250\text{--}333\text{ }\mu\text{m}$	high, but error dependent on accuracy of sample volume and positioning	Suaria et al., 2020
		surface grab	sample bottle, apparatus	all, sample volume constraints may under-represent coarse MP	high, easy to deploy but limited to surface, subsurface	Barrows et al., 2017
		discrete depth grab			high, difficult/time consuming to deploy	Lenaker et al., 2019
		depth/flow integrated			high, difficult/time consuming to deploy	Miller et al., 2021
Snow/ Ice deposit	terrestrial, oceanic	drill/core	sample apparatus	all	high, analysis of different layers possible without cross-contamination	La Khanh et al., 2020
		grab	sample apparatus	all	low, MP concentration too low. Deployed by one study that found only 1 particle per sample typically.	Bergmann et al., 2019

a. Contaminations considerations specific to a given technique in addition to standard field and laboratory contamination sources. b. Range of particle size captured by given technique with notes on potential sampling bias - note that matrices and environmental settings can differ greatly in terms of microplastic (MP) particle size distributions. c. Range of microplastics concentrations or deposition rates observed for a given matrix/technique in previous studies. d. Holistic evaluation of the suitability for each technique/suite of techniques for environmental microplastics pollution sampling.

concentration if paired with appropriate bulk water sampling for calibration and validation (Hitomi et al., 2021; Simmons et al., 2020). Reflectance-based remote sensing of suspended load with aerial and satellite imagery has been used since the late-20th century, with some success for sediments (Warrick and Milliman, 2003; Stumpf and Pennock, 1989; Volpe et al., 2011). Although the detection of microplastics from aerial and satellite imagery is still unfeasible due to image resolution capabilities, measurements of ocean surface roughness by low Earth orbiting bistatic radars have recently been used to detect and estimate marine microplastic concentrations (Evans and Ruf, 2021). Additionally, hyperspectral imagery has been shown to be able to capture microplastic characteristics in samples collected from aquatic and terrestrial environments (Shan et al., 2019; Serranti et al., 2018; Liu et al., 2018) demonstrating the potential for this approach to speed up identification of microplastics in the laboratory. Recently, advances in acoustic (e.g. Acoustic Doppler Current Profilers) and laser-diffraction techniques have permitted the in-situ sampling of concentrations and grain-sizes of natural sediments (Holdaway et al., 1999; Pedocchi and García, 2006; Smeardon and Thorne, 2008; Sassi et al., 2012). Unfortunately, extension of each of these techniques to direct monitoring of microplastics in aquatic systems is unlikely because of the very low concentrations of microplastics relative to limits of detection and the need to distinguish the backscatter signatures of microplastics from that of natural sediment.

Flow integrated sampling (i.e. representatively sampling every part of a channel cross section flow field) has been the 'gold standard' for sediments, although its accuracy is still a challenge (Gitto et al., 2017). Microplastic research would benefit from testing flow integrated sampling strategies in the field, as there are still several issues with current microplastic sampling techniques: a lack of a uniform microplastics sampling has resulted in the propagation of errors into global estimates of microplastic fluxes to the ocean (Weiss et al., 2021). The predominant ocean sampling technique involves trawls with plankton nets (typically 250–330 μm), although this might under-represent the amount of microplastic fibres and particles (Athey and Erdle, 2021). Other techniques involve filtration through a range of different pore sizes, although it is noted that filtration favours the identification of smaller

microplastic fractions, leading to biasing in the reporting of typical microplastic particle sizes found in fluvial and marine environments (Weiss et al., 2021). Notably, microplastic sampling has focussed on surface, or near-surface sampling, yet the wide range of particle size, shape, and density demands characterizing the depth distribution of microplastics concentration to better model transport in both fluvial (Cowger et al., 2021a) and marine (Pabortsava and Lampitt, 2020) environments.

4.1.2. Considerations when sampling for microplastics in the environment

Traditional sediment sampling techniques commonly involve materials or tools that are made from plastic, thus representing potential sources for environmental contamination and should be avoided or sampled to obtain a control sample from it (Woodall et al., 2015). For example, multicoring or piston coring devices, commonly used to sample marine sediments, typically capture sediments in an inner liner made of polyvinyl chloride, which can get damaged when used. Additionally, samples may be exposed to post-collection contamination, through contact with the air and unclean surfaces therefore environmental samples should be sought alongside the use of contamination minimisation procedures. For these reasons it is imperative that field blanks are collected in a manner that effectively represents the magnitude and variability of microplastics contamination introduced through field sampling activities (Brander et al., 2020). The high likelihood of post-sampling contamination can make it challenging to use samples for inquiry outside of plastic pollution research, but careful planning can often permit this.

4.2. Laboratory sampling, identification and classification

Sediments are made up of biotic and abiotic materials (including microplastics). In mineral sediment research, organic material is often removed from the total sediment sample using digestion procedures (Gray et al., 2010) similar to those employed on microplastic samples (Masura et al., 2015). Plastics are generally considered less stable during extraction than mineral sediments due to their low heat tolerance and sensitivity to certain acids and bases (Lusher et al., 2020), which may

discolour them, hence affecting visual observation (Nuelle et al., 2014) or result in fragmentation. The possible fragmentation is important to notice as microplastic particles might change their size or shape during processing (Cole et al., 2013; Krukowski, 1988), which would lead to particle descriptions that differ from the original particle found in the environment. However, it is common to use organic matter digestion (often using a mixture of H_2O_2 and H_2SO_4) for microplastics found in sediments with high organic content before instrumental observations (Cole et al., 2013). Density separation is commonly performed in sediment research to sort sediments by density (Krukowski, 1988) similar to how microplastics (typically of lower density than mineral sediments) are separated from mineral sediments. The fine-grained silt and clay particles that remain adsorbed onto the coarser sediments are removed using dispersants such as sodium hexametaphosphate, which is also the case with microplastics as they may be trapped within the mineral sediments (Pagter et al., 2020). Particle size distribution data for mineral sediments are derived from classical techniques like wet sieving ($>63\text{ }\mu\text{m}$), in conjunction with settling-based techniques such as the pipette method ($<63\text{ }\mu\text{m}$) using Stokes' Law. The different sizes of sediments are isolated by placing the samples on a sieve-stack of different mesh sizes, and then counted under a stereozoom microscope to get abundance data. Similarly, microplastics samples are sieved into size fractions suited for different analytical techniques (i.e. ATR-FTIR vs μFTIR imaging). To obtain quantitative information on the abundance of different sediment grain sizes, instruments such as laser diffraction particle size analyser (LDPSA; range of $0.04\text{--}2000\text{ }\mu\text{m}$) and rapid sediment analysers are used as this is less time-consuming and a large number of samples can be quickly processed unlike the classical techniques. These instruments have been used to study the size distribution of microplastics from commonly occurring plastic particles like polyethylene terephthalate (PET), high density polyethylene (HDPE), polyvinyl chloride (PVC), low density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), and polycarbonate (PC) (Mortula et al., 2021), and personal care products (Renner et al., 2021; Kokalj et al., 2018) obtained from supermarkets. For environmental samples, these techniques are less suited since they cannot distinguish polymers from natural particles. In addition, fluorescent staining with dyes (e.g. Nile Red (Erni-Cassola et al., 2017; Maes et al., 2017)) can be used to distinguish plastic particles from mineral and organic materials in a sample. This method is mainly used for better visualization and subsequent identification of microplastics (Lv et al., 2021). As the staining process with Nile Red depends on the polarity of the plastics, some polymer types (PC, PUR, PET and PVC) and some particle shapes (fibers) are difficult to stain and might thus be overlooked (Prata et al., 2019). Furthermore, natural materials may be co-stained (Prata et al., 2019; Konde et al., 2020). Konde et al. (2020) concluded that photoluminescence spectroscopy can improve the differentiation between plastics and natural materials, while purely optical inspection of the stained samples can lead to misidentification. Fluorescent staining should therefore never be used on its own, but as an additional step to identify microplastic particles in environmental samples.

Shape analysis and observation of surface textural features are often used for sand- and silt-sized sediments. The sphericity and roundness parameters are studied under the microscope using visual estimation (Powers, 1953; Rittenhouse, 1943), and more sophisticated techniques like Fourier transform provides a two-dimensional digitisation of grain images (Lee and Osborne, 1995). The nitrogen gas adsorption method can also be applied to sand-sized mineral particles to derive three-dimensional mineral shape and surface texture (Blott et al., 2004). Advanced techniques such as automatic particle-shape image analysis for natural quartz grains can provide four parameters (solidity, convexity, highly sensitive circularity and aspect ratio), which are used to derive information on the sedimentary processes (Chmielowska et al., 2021). In the case of microplastics, shape classification of larger particles is mainly done with the help of a stereozoom microscope and categorized into foam, pellet, fibre, fragment, and film, with the

measurement of their dimensions (Rochman et al., 2019). Instruments combining laser diffraction and dynamic imaging (e.g. Bettersizer S3 plus; Bettersize Instruments Ltd) can provide information on not only the particle size but also the shape of the material. However, they have not been applied in the case of microplastic studies yet. Recent studies on microplastics have used automated image analysis as a tool to study the dimensions of the plastic particles (Primpke et al., 2019; Primpke et al., 2017; Rodríguez Chialanza et al., 2018), which has helped significantly in reducing the sample processing time. To describe particle shapes with image-based methods, Hentschel and Page (2003) identified that ruggedness (derived from area and perimeter) and elongation (measured by aspect ratio) are the most critical characteristics. Further development of these techniques together with automation of processing will both increase the robustness and speed of microplastic sample processing.

The composition of the mineral particles present in the sediments are studied using their optical mineralogy properties. Scanning electron microscopy-energy dispersive spectroscopy allows a sedimentologist to obtain high-resolution images of the sedimentary particle and also its elemental composition, which aids in the study of surface textural features, and interpretation of their origin and causes. Recent studies on microplastics have also made use of this method to study the effect of environmental degradation processes on the surface texture of microplastics with respect to exposure time (Sait et al., 2021). Destructive and non-destructive techniques are applied to identify the composition of the polymers. Destructive techniques such as differential scanning calorimetry coupled with thermogravimetry (Majewsky et al., 2016) (TGA-DSC), pyrolysis gas chromatography-mass spectrometry (Fries et al., 2013) (Pyr-GC/MS), thermal extraction desorption gas chromatography-mass spectrometry (Dümichen et al., 2017) (TED-GC/MS), x-ray diffraction (Ariza-Tarazona et al., 2019) have been used to this aim. Some of these techniques are also applied to study the composition of organic matter (Ninnes et al., 2017) (Pyr-GC/MS) and mineral fractions (von Eynatten et al., 2012) (XRD) in sediments. Amongst the non-destructive techniques, vibrational spectroscopic methods such as the ATR-FTIR, μFTIR and Raman spectroscopy are the most commonly used to identify the polymer composition, but also nuclear magnetic resonance (Peez et al., 2019) needs to be mentioned as a non-destructive technique. The images obtained by these spectroscopy techniques are also used to determine the 2-dimensional shape of the particles, using automated image analysis. However, limited spectral libraries for material identification are a common problem in microplastics research (Cowger et al., 2021b). Mineralogy research has created a network of labs globally who share Raman and FTIR mineral spectra (<https://rruff.info/>). Pooling resources in this way has also been initiated for microplastic research (<https://simple-plastics.eu/> for IR spectra, SLOPP-E for Raman spectra (Rochman Lab, 2021)), and can be further enhanced in the future.

5. Ecotoxicology

As with natural particles (such as sediment particles) the effects of exposure to microplastics are likely to depend on numerous factors including particle size and shape, chemical composition, and life history of the affected organism. There are drastic differences between the particulate nature of different environmental niches, ranging from relatively particle-free pelagic waters to sediment and soil. Organisms specialized to living in particle-rich environments (such as plants or sediment dwelling invertebrates), or feeding on particles (e.g. filter-feeding crustaceans and bivalves), will be adapted to do so. These organisms, which encounter and ingest non-food particles as a function of their life history (Lopez and Levinton, 1987), employ mechanisms of partition and excretion, which could ostensibly extend to ingesting and excreting microplastics (Gutow et al., 2016). On the other hand, given the ubiquity of microplastics in marine sediment (Woodall et al., 2014), it is likely that many deposit feeding species will encounter and ingest

microplastics to a greater degree than pelagic organisms (Taylor et al., 2016), which may present challenges that go beyond the ingestion of natural particles alone.

In contrast to natural sediment particles, microplastics may contain thousands of harmful chemical contaminants, both as additives and as by-products of their breakdown under environmental conditions (Wiesinger et al., 2021). Wiesinger et al. (2021) determined that more than 10,000 substances are used in the production of plastics, inducing monomers, additives and processing aids, and that more than 2400 of those are substances of concern and potentially hazardous compounds. When plastics are lost into the environment, numerous processes will impact the physical and chemical nature of the materials. As plastics age, chemicals will leach out into the surrounding environment (Hermabessiere et al., 2017) and environmental contaminants will sorb onto the particles (Mato et al., 2001). These chemicals may leach from the microplastics following ingestion, potentially leading to more adverse effects than exposure to relatively more inert natural particles, such as sediments (Zimmermann et al., 2020). However, both microplastics and natural particles may act as vectors for chemical contaminants. Whether microplastics or their associated chemicals impart adverse effects between ingestion and excretion, and whether any effects differ significantly from those of natural particles, remains to be determined for the vast majority of organisms and environmental conditions.

5.1. Interactions with other pollutants and contaminants

The propensity for microplastics to act as chemical vectors has been central to many investigations, but needs to be considered in context and compared to other potential chemical fates and exposure routes. When comparing microplastics and sediments in terms of their interaction with other contaminants, most laboratory studies generally report that microplastics sorb greater or equal amounts of contaminants than soils or sediment particles (Wang and Wang, 2018; Klöckner et al., 2021) with few exceptions. These exceptions are associated with the adsorption of polar contaminants (Zhou et al., 2020a; Besson et al., 2020). Fig. 2 provides a brief overview of microplastic and sediment ranking based on their adsorption capacity to contaminants in laboratory studies. The figure reveals that for sorption of hydrophobic contaminants, sediments consistently rank low compared to plastics while for metals, the reverse is the case. The interaction of any polymer with contaminants is often based on the properties of the polymer, background media or the contaminants (Alimi et al., 2018). Plastic surfaces are often produced to be smooth, hydrophobic and resistant to chemicals (Angu et al., 2014; van Oss, 2020), but these properties can change due to environmental influences such as UV radiation and freezing. Therefore, some microplastics have a relatively smooth and inert surface morphology - leading to a more hydrophobic behaviour - and other

microplastics have rough and charged surfaces, which are more hydrophilic (Hossain et al., 2019). The hydrophobicity is highly dependent on the polymer type and can decrease when a particle gets wet, which is, for example, the case for PE (Švorčík et al., 2006; Simões et al., 2008). The hydrophobic nature of some microplastics (e.g. PVC, PP, PS) make them good candidates for adsorbing persistent organic pollutants in the environment and there is evidence that microplastics in sediment reduce the bioavailability of, e.g. PCBs (Verla et al., 2019), while the general hydrophilic nature of sediments will attract trace metals (Besson et al., 2020; Guan et al., 2020; Qi et al., 2021; Zhou et al., 2020b). The differences in hydrophilicity of plastics and sediment particles might explain why in Fig. 2, sediment particles adsorb metals easily (for example Cs, Zn, Cd) in four studies, but rank low for the three studies investigating hydrophobic contaminants (phenanthrene and triclosan). The importance of microplastics acting as vehicles for contaminant transfer in the ecosystem compared to sediments will be a function of their relative abundances in the environment (Rochman, 2016). Recent estimates show that the current abundance of plastic is significantly less than sediments or other environmental media in the aquatic system, hence, microplastics may not be significant transport vehicles for other contaminants compared to sediments (Koelmans et al., 2016; Gouin et al., 2011).

5.2. Effects in aquatic animals

Despite ample evidence of microplastic uptake across a broad range of aquatic organisms (Duis and Coors, 2016), there is currently little consensus on the ecotoxicological implications, particularly at environmentally relevant concentrations. Much of the early ecotoxicology research into microplastics focused on a few aquatic model organisms, e.g. the planktonic crustacean *Daphnia magna*, and were generally conducted using a limited number of particle types, namely polystyrene spheres (Sá et al., 2018; Wendt-Potthoff et al., 2017) with particle concentrations much higher than found in natural systems (Lenz et al., 2016). Early work addressed several issues, including physiological effects (Wright et al., 2013) and the transfer of chemicals from microplastics to various organisms (Koelmans et al., 2014). Although many studies have demonstrated adverse effects in a plethora of organisms following microplastic ingestion, most studies have neglected to contextualize the impacts of microplastics by including comparisons with other types of particles, such as naturally occurring minerals (e.g. clay and sand) or organic particles (e.g. cellulose fibres and particulate organic matter). This lack of direct comparisons with natural particles means we cannot yet conclude that the toxicological effects of microplastics extend beyond those of natural particles. Indeed, a high concentration of natural particles such as silt and sediment is a known stressor for many species (Österling et al., 2010), and the effects of

	PE	PA	PS	PP	PVC	PET	S	Contaminants
Teuten et al., 2007	1			2	3		4	phenanthrene
Wang and Wang, 2018	1		2		3		4	phenanthrene
Chen et al., 2021	1		2				3	triclosan
Besson et al., 2020	2						1	caesium, chromium, zinc
Guan et al., 2020			2				1	cadmium, cobalt, copper, nickel, silver, zinc
Qi et al., 2021			2				1	lead
Zhou et al., 2020		2	4		3	5	1	cadmium

Fig. 2. Comparison of the interaction of microplastics and sediments with contaminants. A value of 1 is assigned to the particle (plastic or sediment) with the highest sorption capacity reported in a given study and increasing values are for particles with lower sorption capacities in the same study. PE - polyethylene, PA - polyamide, PS - polystyrene, PP - polypropylene, PVC - polyvinyl chloride, PET - polyethylene terephthalate, S - Sediment. Colours are only used to differentiate visually between ranking numbers. References: Wang and Wang, 2018; Besson et al., 2020; Guan et al., 2020; Qi et al., 2021; Zhou et al., 2020b; Teuten et al., 2007; Chen et al., 2021

turbidity due to suspended sediment in aquatic ecosystems are generally well-understood (Henley et al., 2000).

In recent years, several studies have attempted to disentangle the toxicological effects of microplastics from those of natural particles, by comparing both particle types to particle-free controls. Using this approach, several adverse effects have been recorded in aquatic animals in response to microplastic exposure including reduced growth and development, reduced fecundity, increased mortality, and adverse generational effects (Schür et al., 2020; Scherer et al., 2020; Zimmermann et al., 2020; Casado et al., 2013; Puranen Vasilakis, 2017). In contrast, exposure to natural particles under the same experimental conditions has been shown to have little to no effect. However, most of these studies have used *D. magna* as the model organism, and so the external validity of these results to other species and systems is unknown. It is also important to note that a control, such as kaolin may not be suitable for the assessment of microplastic toxicity, given its propensity for sedimentation, possibly resulting in lower bioavailability.

It is not yet clear if gut retention time for sediment versus microplastics is different across different taxa. For example deposit feeding benthic organisms rely on the organic fraction of ingestion sediment for their nutrition (Lopez and Levinton, 1987). Given that microplastics are ubiquitous in aquatic sediments (Woodall et al., 2014) these deposit feeders are interacting with microplastics as they feed, and many studies have documented internalization of microplastics in this type of organism (Taylor et al., 2016). Exposure to high levels of microplastics can be impactful to deposit feeders (i.e. by reducing gut activity (Wright et al., 2013)) and could result in the reduction of inorganic nutrient release (Green et al., 2016), thereby impacting sediment-based processes.

5.3. Effects in plants and algae

Both natural particles and microplastics may adversely affect plants, depending on particle properties. In the case of microplastics, exposure may affect growth and germination (Bosker et al., 2019; Zhang et al., 2021b), ostensibly caused by the release of chemical additives such as flame retardants and plasticizers (Hahladakis et al., 2018). Conversely, the presence of natural particles has been found to exert little to no effect on plant growth (Rozman et al., 2021; Moore et al., 2021; Kiani et al., 2021), even stimulating growth in algae in some cases (Gorokhova et al., 2020).

A small number of studies have directly addressed the effects of microplastics versus natural particles, using aquatic plants and algae. Rozman et al. (2021) showed that irregularly shaped microplastics and Bakelite (a thermoset plastic, developed at the beginning of the 20th century) negatively affected growth in the duckweed *Lemna minor*, with Bakelite apparently leaching inhibiting chemicals. In contrast, natural particles did not impart any negative effects. However, Gorokhova et al. (2020) found that both microplastics and natural particles reduced growth in the unicellular alga *Raphidocelis subcapitata*, with the greatest inhibition occurring at the highest concentrations. Again, this variation in effect may be related to life history differences between macro- and microalgae. It is also possible that the deposition of microplastics, as with natural particles, contributes to the inhibition of plant growth (Brodersen et al., 2017).

5.4. Effects on microbes

Microbial communities have been found on microplastics and in sediment, and microbes are vital in many planetary processes, ecologically acting as producers and decomposers. It is still uncertain how benthic microbial communities that reside on these different particles differ, however there is increasing evidence that microplastics harbour microbes that are significantly different to those of surrounding sediment (reviewed by (Yang et al., 2020)). In addition, a recent study showed that microplastics altered the microbial community and

nitrogen cycling process of marine sediment of a mesocosm experiment (Seeley et al., 2020). This demonstrates the close link between microplastic presence and the ecological function of sediment communities.

5.5. Variation in effect based on particle properties

Controlled studies suggest that the type of particle and its source may affect toxicity. Ogonowski et al. (2016) showed that secondary microplastics (cryomilled PE, irregular shapes, avg. 2.8 µm) increased mortality, reduced fecundity, and delayed reproduction in *Daphnia magna*, while primary microplastics (PE spheres, avg. 4 µm) and kaolin (irregular shapes, avg. 4.4 µm) imparted no significant effects. Zimmermann et al. (2020) exposed *Daphnia magna* to cryomilled, secondary microplastics, <59 µm which is small enough to be consumed by animals, and found that polyvinyl chloride (PVC) reduced reproduction, while polylactic acid (PLA) reduced survival. The authors had exposed the *Daphnia* to microplastics grinded from consumer products which can contain thousands of chemicals, so they included several different exposures to differentiate between chemical and particle effects: produced microplastics, microplastics that had been purified from chemical additives via methanol extractions, and the chemical extracts. Further analysis showed that the effects of PLA were caused by the physical properties of the microplastics, while the effects of PVC were chemical in nature. In contrast, Puranen Vasilakis, 2017 showed no significant effect related to PLA exposure, but negative effects in response to polystyrene were shown, ostensibly caused by the release of styrene monomers from the latter. In relation to natural particles, differences in toxicity between kaolin and diatomite have recently been shown (Scherer et al., 2020). This suggests that some natural particles, while apparently more benign than microplastics, may also exert negative effects in aquatic organisms. Again, further testing is required to determine the effect thresholds for different natural particles, as well as microplastics.

5.6. Meta-analysis of direct ecotoxicology comparisons between different particle types

We have extracted data from studies that compare the different effects of exposure to microplastics versus natural particles (Schür et al., 2020; Scherer et al., 2020; Zimmermann et al., 2020; Puranen Vasilakis, 2017; Rozman et al., 2021; Gorokhova et al., 2020; Ogonowski et al., 2016), to differentiate between particle driven impacts and those inherent to microplastics. We only included studies that had a negative, particle-free control, i.e. aqueous solution to which neither sediment nor microplastic particles have been added. The forest plots in Fig. 3 show the effects of natural particles and microplastics on growth, reproduction, and mortality/survival of several aquatic animal and plant species. Values to the left of the line indicate negative effects (lower growth, higher mortality etc), while values to the right of the line indicate positive effects. Values at the line indicate no effect. As such, greater distance from the line represents a greater effect magnitude. The results suggest that exposure to low concentrations of microplastics or natural particles do not affect growth, reproduction, or survival in the analysed species. At higher concentrations, microplastics may exert negative effects on reproductive output and survival, while high concentrations of both microplastics and natural particles may adversely affect growth. However, given the low number of studies and the high degree of heterogeneity, the results must be interpreted with caution.

6. Conclusions and recommendations

We have compared the particle properties, aquatic transport dynamics, environmental sampling and ecotoxicology of microplastics and natural sediments. While knowledge on natural sediments is already broadly used in some areas of microplastics research (transport behaviour, environmental sampling), other areas may benefit from the transfer of sedimentological knowledge to microplastics (particle

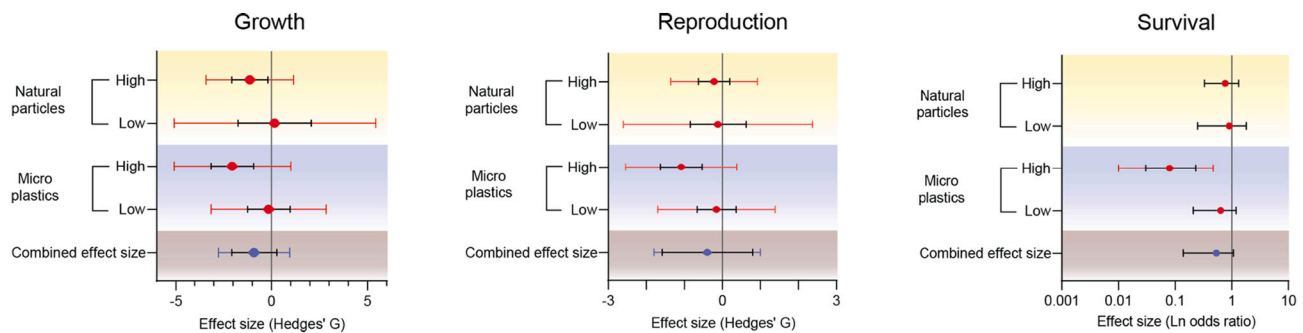


Fig. 3. Results of a meta-analysis of Schür et al., 2020; Scherer et al., 2020; Zimmermann et al., 2020; Puranen Vasilakis, 2017; Rozman et al., 2021; Gorokhova et al., 2020; Ogonowski et al., 2016 to assess the effect of high and low concentrations of both natural particles and microplastics on aquatic biota. The analysis can be requested from the corresponding author. This analysis grouped effects on both animals and plants, given the small number of studies for each response ($n = 4-7$). Confidence and prediction intervals (95% in both cases) indicate that microplastics may affect growth, reproduction, and survival at higher concentrations, relative to natural particles. However, given the high degree of uncertainty, more data is required to confirm this.

description) or from acknowledging the differences and similarities between sediments and microplastics (numerical modelling, ecotoxicology). Based on these insights, we synthesised a number of important areas for future research (Table 3).

Many studies have suggested that the paradigms used in sedimentary research can be applicable to describing the fate of microplastics, as they can be regarded as a type of sediment that behaves in similar ways, with erosion, transport and deposition depending on grain size. However, microplastics are diverse and characterized by different densities, particle sizes and shapes, as opposed to natural sediments that have successfully been generalized to be spherical and uniformly size-distributed in the past. This generalization does not seem to be suitable for microplastics due to their greater shape diversity as described above. Therefore, we need to improve and standardize our description of microplastic particles to gain insights into how particle properties influence the distribution and ecotoxicology of microplastics in the environment. Moreover, a standardized property description might allow us to determine particle origin. That is why methods for particle description in sedimentology, such as the use of shape descriptors or the principle of the cube, can be useful and should be adopted by microplastic researchers. In addition to the high particle variability of microplastics, their change over time complicates our understanding of their behaviour and predictability. Factors such as changes in particle shape through fragmentation, degradation and deformability that received little attention in the field of sediments may be essential for microplastics behaviour and require further research.

The classification of sediment transport into suspended and bedload transport has already been applied to microplastics research and expanded to a third class, i.e. surface transport. Especially bedload transport suffers from many open research questions, such as the conditions that lead to near-bed particle movement, the quantities of transport and particle interactions with the bed. Shifts between these transport modes that usually can be estimated by particle size and density of natural sediments are still poorly understood for microplastics and require the development of metrics that relate particle properties with transport mode.

The possibilities of numerical simulations of sediment transport have not yet been fully exploited for microplastic research, although basic paradigms from sediment transport modelling exist that can be of use for the growing field of microplastic modelling. Describing microplastic particle transport based on sedimentary laws provides important insights for global microplastic distribution, but lacks the consideration of the dynamic behaviour of the microplastics themselves. To differentiate the behaviour of natural and microplastic particles, we need to parameterize microplastics' physical properties, shape, surface properties, density, as well as time-dependent changes, including biofouling, aggregation, flocculation and fragmentation. In the future, numerical

simulations should consider the above-mentioned findings, or emphasize the assumptions of their models more clearly.

In particular the role of biota in microplastic transport requires more research. While vegetation effects are well-studied for natural sediments, few studies quantify the mechanisms behind an abundance of microplastics in vegetated surfaces. Similarly, alterations of resuspension and deposition thresholds induced by biofilms and macrobenthic organisms need to be quantified. Here, our knowledge from sediment resuspension appears to be applicable and representative of microplastics behaviour. Conducting flume experiments similar to those in sedimentary research can be useful to determine threshold shear stresses for microplastic resuspension, which we urgently require to incorporate microplastics in numerical models and test hypotheses on the processes behind microplastic redistribution.

Due to the complex nature of sediments and microplastics, existing methodologies for environmental sampling are highly varied and not immediately comparable, highlighting the need for standardized guidelines for future sediment and microplastic studies. This involves the entire process of the assessment of microplastics in environmental samples, from sampling technique, sample preparation and identification of microplastic particles. The selection of an adequate sampling technique is a vital step in environmental analysis and the overview here, including notes on the suitability of current methods for microplastic sampling, can be used as a useful tool in creating standardised guidelines. Special attention should be paid to the prevailing modes of microplastic transport in order to carry out representative measurements.

Much work has been done to understand the impacts of microplastics on organisms, mechanisms of interactions and uptake, physical damage, and important factors driving toxicity. This research allows us to begin to understand the environmental risk posed by these complex particles. However, we are still grappling to fully comprehend the relative importance of the polymers themselves, the plastic-associated chemicals and the physical nature of the particles. Experimental design and choice of particle exposures is therefore important, natural particles should be included in future toxicity studies, whether the work aims to understand chemical vector effects or particle effects, so that we might gain perspective and better understand relative risk.

In our review, we highlight that the profound knowledge regarding particle description, transport dynamics, sampling methods and ecotoxicology of sediments can be of great value for microplastics research and has already offered valuable concepts to describe microplastics behaviour and transport. It would be foolhardy not to take advantage of the opportunities that arise from the legacy of sediment research to guide future microplastic research. Nevertheless, caution is advised, as the transferability of sedimentary methods and principles must first be assessed and, if necessary, adapted to microplastics in order to be

Table 3

Open research questions. The interdisciplinary comparison of microplastics with natural sediment revealed a variety of knowledge gaps from which future research goals (RG) can be derived. Priority research areas are listed here, along with potential knowledge benefits, and specific tasks to be achieved.

Research goal	Benefits	Tasks
RG1. To improve and standardize descriptions of microplastic particles	This will allow comparability of individual studies and to understand the impact of the particle properties on the transport and the ecotoxicology of the particles	1. Define particle size and principal dimensions of non-spherical particles (e. g. by using shape descriptors from sedimentology or by sedimentological methods such as the 'principle of the cube') 2. Assess the suitability of currently used shape categories and building on this to develop new shape categories when necessary 3. Describe and parameterize the impact of different shapes and deformability of microplastic particles on their transport behaviour, which highly distinguishes microplastics from natural sediment 4. Investigate the implications of microplastic density being close to that of water, making it easy for the particles to move from a buoyant to settled state
RG2. To understand and quantify time-variable particle property changes and interactions with other environmental substances and their impact on microplastic transport	This will allow implementation of time-dependent changes into numerical models and improve our understanding of the impact of those changes on microplastic transport	1. Investigate time-dependent changes of particle properties, especially during processes of biofouling, degradation and fragmentation, and parameterize their impact on transport behaviour 2. Investigate interactions of microplastics with other environmental substances, e.g. aggregation and flocculation, and parameterize their impact on transport behaviour
RG3. To evaluate the vertical distribution of aquatic microplastic and their transport in the water column	This will enhance our understanding of microplastics transport behaviour and improve the reliability of environment monitoring	1. Based on the concepts used in sediment transport, quantify the distribution of the total microplastic transport as surface, suspended, and bedload transport and describe the mechanisms behind bedload transport of microplastics
RG4. To evaluate differences in the erosion and deposition behaviour between microplastics and sediments	This is imperative to permit our understanding of microplastics retention, remobilization, flux and accumulation, and resulting transport behaviour	1. Quantify the influence of particle properties (e.g. shape, surface properties) on the onset of motion 2. Determine resuspension of particle mixtures and particle densities, both for microplastics-microplastics and microplastics-sediment mixtures

Table 3 (continued)

Research goal	Benefits	Tasks
RG5. To understand and quantify the impact of biota on microplastic transport	This will enhance our understanding of microplastic deposition and transport in different ecosystems, depending on the presence of vegetation and/or benthic organisms	1. Compare vegetation-induced microplastic settling with that of natural sediments 2. Quantify the effect of biota (including biofilm) on resuspension thresholds and bedload transport
RG6. To improve and standardize sampling methods	This will provide more robust and detailed descriptions of the occurrence and types of microplastics in the environment and improve the comparability of results from different studies	1. Develop refined techniques and parameters for sample collection, preparation and analysis, taking microplastics transport mechanisms into account 2. Assess the application of sedimentological methods such as flow-integrated sampling or develop innovative techniques such as remote sensing, hyperspectral imaging, acoustics or laser diffraction methods to microplastics research
RG7. To study the drivers of microplastic toxicity in comparison with sediment particles	This will improve knowledge of the relative importance of the polymers themselves, the plastic-associated chemicals and the physical nature of the particles for their ecotoxicology compared to other particles in the environment	1. Improve experimental design and toxicity testing by inclusion of natural particles that are similar to the MPs under investigation

applied effectively.

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Declaration of Competing Interest

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

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