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Abstract

Background

Diets rich in dietary fibre are associated with multiple health benefits, but there is often only a restricted understanding of the mechanisms underlying these associations. This limits the ability to select or design foods for specific nutritional purposes. Traditionally, the diverse physical and chemical forms of dietary fibre have only been categorised as either soluble or insoluble.

Scope and Approach

In this review, the physicochemical properties that have been proposed to be responsible for the biological functionality of dietary fibres in the digestive tract are summarised and classified. The extent to which these properties follow naturally from categorisation into soluble vs insoluble forms are then assessed. Based on this analysis, a new approach to functional categorisation of dietary fibres is proposed.

Key Findings and Conclusions

The physicochemical properties of dietary fibre components that are relevant to digestive tract functionality can be classified under the headings of binding, structuring, and transport barriers. Major nutritional outcomes such as control of macronutrient digestion or the nature of residual digesta that are available for fermentation by the large intestinal microbiota depend on combinations of these physicochemical properties in ways which are not readily reflected by a soluble vs insoluble fibre definition. An alternative approach is proposed based on 2D mapping of dietary fibre materials as a function of molecule/particle size and local density. This effectively separates diverse fibre materials and can be linked semi-quantitatively with functionally-important properties.

Functional Categorisation of Dietary Fibre in Foods: Beyond 'Soluble' vs 'Insoluble'

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1. Introduction

1.1 Dietary fibre intake is associated with good health outcomes

Results from a number of large prospective cohort studies have shown clear associations between dietary fibre intake and reduced risks of all-cause mortality, cardiovascular diseases, diabetes and cancers of the digestive tract (Anderson et al, 2009; Chuang et al, 2012, Threapleton et al, 2013). As most dietary fibre is in the form of foods derived from cereals, fruits, vegetables, legumes and nuts, this is reflected in consensus health advice around the world that a diet rich in plant-based foods provides the best dietary protection against non-communicable diseases. Some studies have attempted to identify specific protective effects of fibre from each of cereals, vegetables and fruits. This is more challenging because most people eat all three food types, but the analysis to date suggests that there may be some differences between these broad classes, with cereal fibre being particularly protective (Park, Subar, Hollenbeck & Schatzkin, 2011; Huang, Xu, Lee, Cho & Qi, 2015; Aune et al, 2016). Whilst these epidemiological studies can be statistically powerful and have a place in deriving population-level dietary guidelines, they show correlations not causations. It is therefore frequently identified that greater mechanistic understanding of the protective actions of fibre is needed in order to provide more tailored dietary advice and guide the design of formulated food with optimised nutritional benefit (Chuang et al, 2012; Gidley 2013; Jones 2013; Grundy et al, 2016; Capuano, 2017).

Hypotheses for the protective action of at least some fibres against diabetes, cardiovascular disease and colon cancer have been proposed (Gidley 2013; Jones 2013), with a focus on carbohydrate (diabetes) and lipid/sterol (cardiovascular disease) metabolism, food intake limitation (satiety), and/or large intestinal microbiota (colon cancer). However, there is a large gap between whole-of-diet data, analysed at the population level to derive correlations,

and mechanistic studies that typically focus on single ingredients. One of the challenges in bridging this gap is the lack of a coherent framework for connecting relevant measurable properties of specific fibre components with the mechanisms by which they may influence health outcomes as diverse as microbiome modulation, nutrient uptake rates, gastrointestinal passage rates, and satiety.

1.2 Dietary fibre is structurally and functionally diverse

Dietary fibre in foods ranges from intact plant tissue pieces to small oligosaccharide molecules. The boundaries of what is 'in' or 'out' of a definition of dietary fibre have been debated for decades, but a consensus is now forming around a definition adopted by CODEX in 2009. This definition is based on carbohydrate polymers that are not hydrolysed by the endogenous enzymes in the small intestine of humans and are either (i) naturally occurring in food, (ii) obtained from food raw materials by extraction, or (iii) synthetic carbohydrate polymers. The key difference between type (i) and types (ii) and (iii) are that the latter are qualified to only include those materials "which have been shown to have a physiological effect of benefit to health as demonstrated by generally accepted scientific evidence to competent authorities" (Jones, 2013). Thus, a clear distinction is drawn between endogenous and extracted/synthetic carbohydrate polymers, which is consistent with health agency dietary guidelines (based on prospective cohort studies) that focus on natural foods such as whole grains, vegetables and fruit at the expense of those foods which are based on recombination of refined ingredients. There are a number of questions of inclusion and exclusion surrounding the CODEX definition. One is the minimum size (degree of polymerisation; DP) which was initially set at DP10 with the option for individual countries to reduce this to DP3. From a scientific and practical perspective (Jones, 2013), it seems likely that DP3 will become the de facto standard. A second area is the lack of explicit inclusion of lignin, which is not a carbohydrate polymer but is an intrinsic (but usually minor)

component of many edible plant tissues, and may contribute to health-related functional properties.

Within each of the three broad classes of natural, extracted, and synthetic dietary fibres, there is great structural diversity at the chemical as well as the physical structure level. Categorisation of fibres in terms of chemical composition is often used. This has the benefit of the analytical methods being robust, accurate and repeatable, but has major drawbacks in that it does not usually include molecular size characterisation (so an oligosaccharide is treated as equivalent to a polysaccharide), and does not usually distinguish between isolated molecules and those which are part of a natural matrix, such as plant cell walls. The natural heterogeneity of intrinsic fibre in plant-based foods at the polymer, cell wall, and tissue level (Burton et al 2010) also provides many challenges in generating a sufficiently complete molecular characterisation of dietary fibre components to address issues of nutritional functionality.

In addition to diversity at the structural level, there is apparent diversity in the mechanisms underlying nutritional functionality at all stages of digestive processing that make it challenging to relate food composition to potential health outcomes (Capuano, 2017). For example, the oral breakdown of solid plant-based foods through mastication can boost the liberation of starch and/or sugar and thereby influence the rate of glucose absorption into the blood (Ranawana, Monro, Mishra & Henry, 2010). Another example is the structuring properties of dietary fibres that through modulation of rheological (flow) properties can influence gastric residence time and therefore impact satiety as well as nutrient absorption (Mackie, Bajka & Rigby, 2016). In addition, fibre components can bind or encapsulate micronutrients controlling their bioaccessibility and hence modulate their bioavailability (Padayachee et al, 2017). Finally, the rate of passage of digesta in the small intestine can be increased by dietary fibres, potentially resulting in delayed nutrient uptake and the triggering

of the 'ileal brake' (van Avesaat, Troost, Ripken, Hendriks & Masclee, 2015), as well as affecting the hydration of large intestinal contents, as exemplified by the faecal bulking effect, which is greater for complex vegetable tissues than more refined fibres (Monro, Mishra, Redman, Somerfield & Ng, 2016). By definition, dietary fibres are not digested by human enzymes in the stomach or small intestine, and are therefore transported to the large intestine where they can act as an energy source for the resident microbiota together with any co-passenger micro- and macronutrients (Padayachee, Day, Howell & Gidley, 2017; Dhital, Warren, Butterworth, Ellis & Gidley, 2017). The rate at which this fermentation occurs can vary from very fast (with consequent potential for gastrointestinal discomfort) to very slow (with consequent excretion of much of the fibre), largely dependent on the physical structure of the digesta. The consequences for microbiome populations will also vary with fibre type, but this is more likely to be due to chemical composition as specific microbial community members can contribute the range of hydrolytic activities required to degrade specific polysaccharide structures.

1.3 Solubility is a limited indicator of dietary fibre functionality

Apart from chemical structure, the other characteristic that has been traditionally used to describe dietary fibre types is solubility. Typically, fibre solubility is evaluated after a food or component has been digested under conditions related to those found in the gastrointestinal tract (McCleary et al, 2012) and is separated from insoluble fibre by filtration or centrifugation. Sometimes, solubility is assessed prior to in vitro digestion, and the temperature regimes, centrifugation speeds or filtration cut-offs are often not standardised. Nevertheless, there are clear examples of soluble fibres such as many low molecular weight oligosaccharides and some polysaccharides, and similarly obvious insoluble fibres such as cereal brans, fruit and vegetable skins. However, there is a large number of dietary fibre types found in foods which either have elements of both soluble and insoluble fibre (e.g. cereal

126 flours; Comino, Collins, Lahnstein, Beahan & Gidley, 2014), or which have highly hydrated
127 but insoluble forms (e.g. fruit and vegetable purees; Padayachee, Day, Howell & Gidley,
128 2017). This range of solubilities creates a physical continuum stretching from easily soluble
129 fibres, to poorly soluble, swollen gel-like networks through to insoluble fibres.

130 Various nutritional functionalities are traditionally ascribed to either 'soluble' or 'insoluble'
131 fibre. Soluble fibre is often reported as increasing the 'viscosity' of digesta with consequent
132 effects on reducing gastric emptying and slowing nutrient absorption. The digesta flow
133 profile (rheology), however, depends on the applied stress, and can be shear thinning or
134 exhibit a yield stress behaviour (Lentle and Janssen, 2010). In the case of the former,
135 viscosity (resistance to shear deformation) reduces with applied stress, while in the case of
136 the yield stress fluid the onset of flow ('yield') occurs only above a critical value of the stress
137 ('yield stress'). In the context of foods, shear thinning is typically associated with high
138 molecular weight polymers in solution and yield stress behaviour with networks of food
139 particles. For dietary fibres, the 'viscosifying' effect thus can be due to increase in viscosity
140 or viscoelasticity for high molecular weight soluble polysaccharides, but also for hydrated but
141 insoluble materials such as oat bran or fruit and vegetable fibres, where yield stress behaviour
142 can emerge. Conversely, low molecular weight soluble fibres such as oligosaccharides would
143 not be expected to have any direct 'viscosifying' effect.

144 Further, digesta can demonstrate a significant degree of viscoelasticity (Shelat et al, 2015),
145 defined as the ratio of the loss modulus (viscous part) to the storage modulus (elastic part), as
146 well as exhibiting non-linear rheological effects which result in deviations between shear,
147 squeeze and extensional deformations, which are all present during gastrointestinal transit
148 (Lentle and Janssen, 2010).

149 The effect of comminution e.g. particle size reduction as a result of oral, gastric or intestinal
150 processing, is related to the ability of particles to adhere to each other. These interactions are
151 frequently driven by capillary forces, for example due to the presence of microscopic gas
152 bubbles, as well as due to interaction and bridging adhesion between surface polymer layers
153 of insoluble particles. The adhesive interaction promotes particle clustering resulting in the
154 formation of a cohesive semi-solid. Insoluble fibre, such as cereal brans and seeds, which
155 absorb water and form a polymer-rich interfacial layer can facilitate comminution, and are
156 often described as having the ability to promote the softening of digesta and support regular
157 bowel movements. By contrast, highly condensed or lignified tissues such as cereal hulls or
158 leaf stalks would not be expected to absorb water and thus would display weak adhesive
159 interactions that limits their ability to facilitate comminution.

160 An over-simplification that is sometimes made is that soluble fibres are readily fermented by
161 the resident microbiota but insoluble fibres are not. There are, however, many examples of
162 insoluble fibres e.g. from fruit, vegetable or cereal sources that are readily fermented, and
163 there are a few examples of soluble fibres whose chemistry is apparently so complex that
164 microbial enzymes are unable to hydrolyse them significantly (e.g. psyllium and other
165 mucilage gums). For the case of cereal flours and derived foods, it has recently been shown
166 that there is very similar fermentation behaviour for soluble and insoluble fibre fractions with
167 similar chemical compositions (Comino et al, 2018)

168 There is clearly a need for a more sophisticated way of categorising dietary fibres that is
169 linked to their nutritional functionality, as neither chemical composition nor fibre solubility
170 are sufficiently discriminatory.

171 *2. Dietary fibre functionality is linked to structuring, binding and/or barrier properties*

The main nutritional functionality of dietary fibres can be simplified to effects in the digestive tract on:

- nutrient digestion and uptake rates
- residence times and passage rates
- fermentation products and microbiota populations,

but each of these is influenced by many different fibre physicochemical properties. For example, nutrient digestion and uptake rates may be influenced by structuring effects that limit the access of digestive enzymes to macronutrient substrates (protein, triglyceride, starch) or the transport of hydrolysed products to the epithelial cell layer, where fibre effects on the mucus may attenuate uptake (Mackie, Bajka & Rigby, 2016; Capuano, 2017). Alternatively, macronutrient digestion may be limited by encapsulation within plant cellular structures (Grundy et al, 2016) and food gels or by complexation with other food components in condensed forms (Zhang, Dhital & Gidley, 2015) e.g. starch in wholemeal pasta. These multiple approaches to achieving comparable outcomes suggest that there are underlying properties that are more characteristic of individual fibre materials. We suggest that these comprise:

- bulk structuring
- molecular binding
- transport barriers

Bulk structuring effects of a fibre relate to digesta rheology once interactions with other components are taken into account, and are expected to influence e.g. digesta passage rate, enzyme digestion rates, nutrient transport and fermentation kinetics. Molecular binding of fibres with enzymes (Dhital, Gidley & Warren, 2015), micronutrients (Padayachee, Day, Howell & Gidley, 2017), bile salts (Gunness, Flanagan, Mata, Gilbert & Gidley, 2016),

196 mucins (Mackie, Bajka & Rigby, 2016; Sriamornsak & Wattanakorn, 2008; Meldrum,
197 Yakubov, Gartaula, McGuckin & Gidley, 2017) and bacteria (Gorham, Williams, Gidley &
198 Mikkelsen, 2016), as well as with other food components, are an under-appreciated feature of
199 many polymeric and particulate dietary fibres. These effects can contribute to all aspects of
200 digestion, passage and fermentation through e.g. reducing enzyme activities, preventing
201 micronutrient bioaccessibility, limiting absorption processes and affecting microbial
202 fermentation. Transport barriers act to separate micro- or macronutrients from other digesta
203 components and typically involve a locally dense structure that is sufficient to limit molecular
204 transport. Examples include encapsulating systems such as plant cells (Dhital, Bhattarai,
205 Gorham & Gidley, 2016) and food gels or condensed processed food forms such as
206 wholemeal pasta (Zou, Sissons, Warren, Gidley & Gilbert, 2016).

207 Whilst structuring, binding and barrier properties provide a reasonably comprehensive
208 framework for categorising the physicochemical properties important for nutritional
209 functionality of dietary fibres, this does not lead directly to classification of the properties of
210 individual types of fibre. For this, the characteristic structural features of fibres that
211 contribute to structuring, binding and barrier properties need to be identified.

212 Structuring (rheology) of dietary fibres in digesta can come from both soluble polymers and
213 swollen particles with both polymer/particle size and concentration being key determinants.
214 Binding phenomena will be expected to involve some specific chemical features. For
215 example the negative charge of pectins serves to enhance binding with positively charged
216 mucin (Sriamornsak & Wattanakorn, 2008; Meldrum, Yakubov, Gartaula, McGuckin &
217 Gidley, 2017) or anthocyanin (Phan, Flanagan, D'Arcy & Gidley, 2017), but reduces binding
218 with phenolic acids (Phan, Flanagan, D'Arcy & Gidley, 2017). More generally, local
219 molecular rigidity and/or density of fibre polymers should be expected to enhance binding
220 through presenting a structurally consistent surface. The key to maintaining an efficient

transport barrier is to reduce the effective pore size such that e.g. digestive enzymes are retarded or prevented from crossing it. Both molecule/particle size and local density are therefore important considerations. Based on this analysis, molecule/particle size and local density/concentration are the key characteristics of fibre components that would be expected to be related to structuring, binding and barrier properties and therefore to nutritional functionality.

3.1 Mapping dietary fibres as a function of size and local density

We propose that a useful approach to categorising the physicochemical properties of diverse types of dietary fibre is to map them by their size and their local density under application conditions, typically fully hydrated. A size axis can cover both dissolved molecules (hydrodynamic size) and particles, with dimensions ranging from about 1 nm for a trisaccharide (the smallest molecule that can be classified as dietary fibre) up to the mm/cm scale for large pieces of cereal bran or fruit/vegetable pulp. Although bulk concentration could be used as another axis, it is argued above that local concentration or density is a more appropriate measure for determining both binding and transport properties. Of course, for dissolved molecules, concentration and local density are equivalent, it is only for particulate materials that the two measures diverge. The range of concentration or density can range from a practical lower level of about 0.1 g/100g up to highly condensed systems at close to 100g/100g. For both size and concentration/density, the wide range of possible values suggests that a logarithmic rather than a linear scale would be appropriate for each axis. Such a plot is shown in Figure 1, populated by selected examples of dietary fibre types. Apart from the top left hand corner (which is bound by the physical solubility limit of oligo- or polysaccharides), essentially the whole of the area in Figure 1 is sampled by different dietary

fibre types under realistic food and digestion conditions, giving the potential for a high level of differentiation between individual dietary fibres. We note also that during oral or digestive processing of food both size and local density may be altered by mechanical or (bio)chemical conditions, allowing the possibility of tracking changes across the plot illustrated in Figure 1.

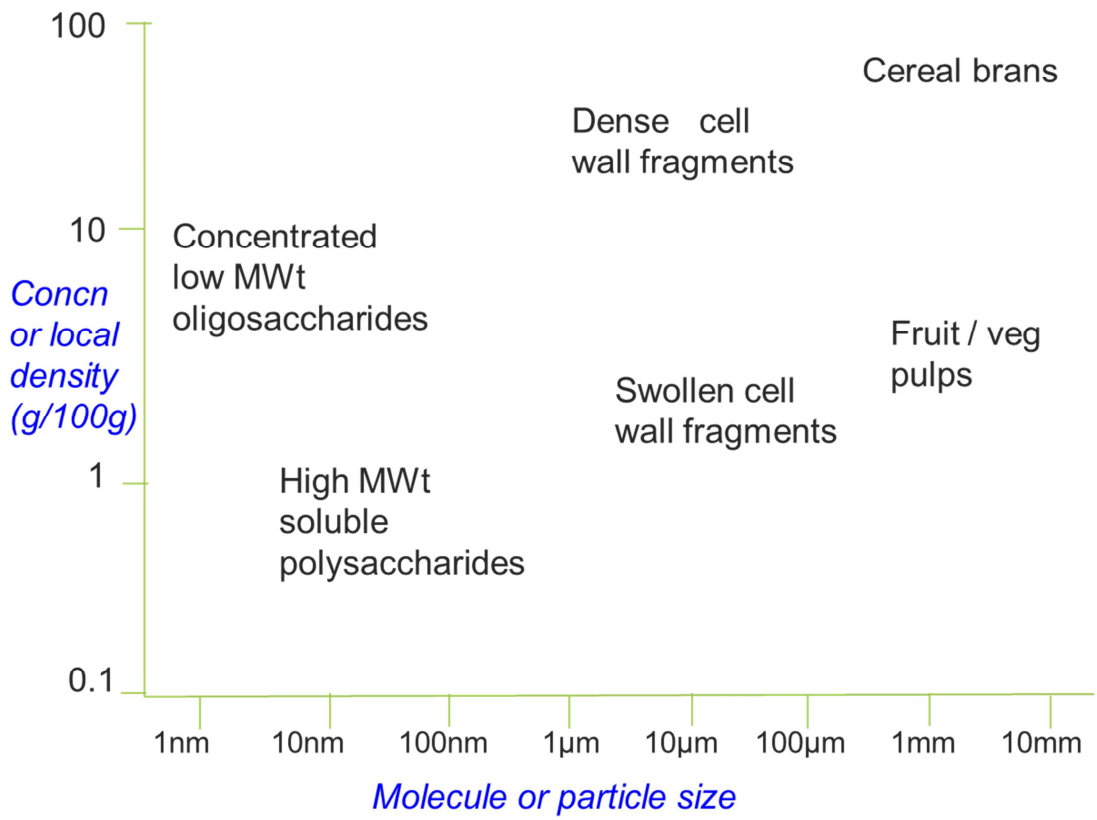

Figure 1. Mapping of example types of dietary fibres against their molecular or particle size and concentration or local density. Positions of fibre types are illustrative and not intended to be quantitatively precise.

3.2 Size / density plots allow differentiation of solubility, viscosity, binding and fermentation properties

To test the utility of size/density plots (Figure 1) to distinguish between fibre properties, approximate boundaries between soluble/insoluble, 'flowing'/non-'flowing',

limited/extensive binding, and rapid/slow fermentation behaviours are illustrated schematically in Figure 2.

Solubility is limited by the size of polymers/colloids that can dissolve. This is in the 100 nm – 1 µm range for polysaccharides in water, above which entities would normally be expected to phase separate. As the conventional soluble fibre test is carried out under dilute conditions, the starting concentration would not be expected to influence the solubility markedly – hence the vertical boundary division (Figure 2A).

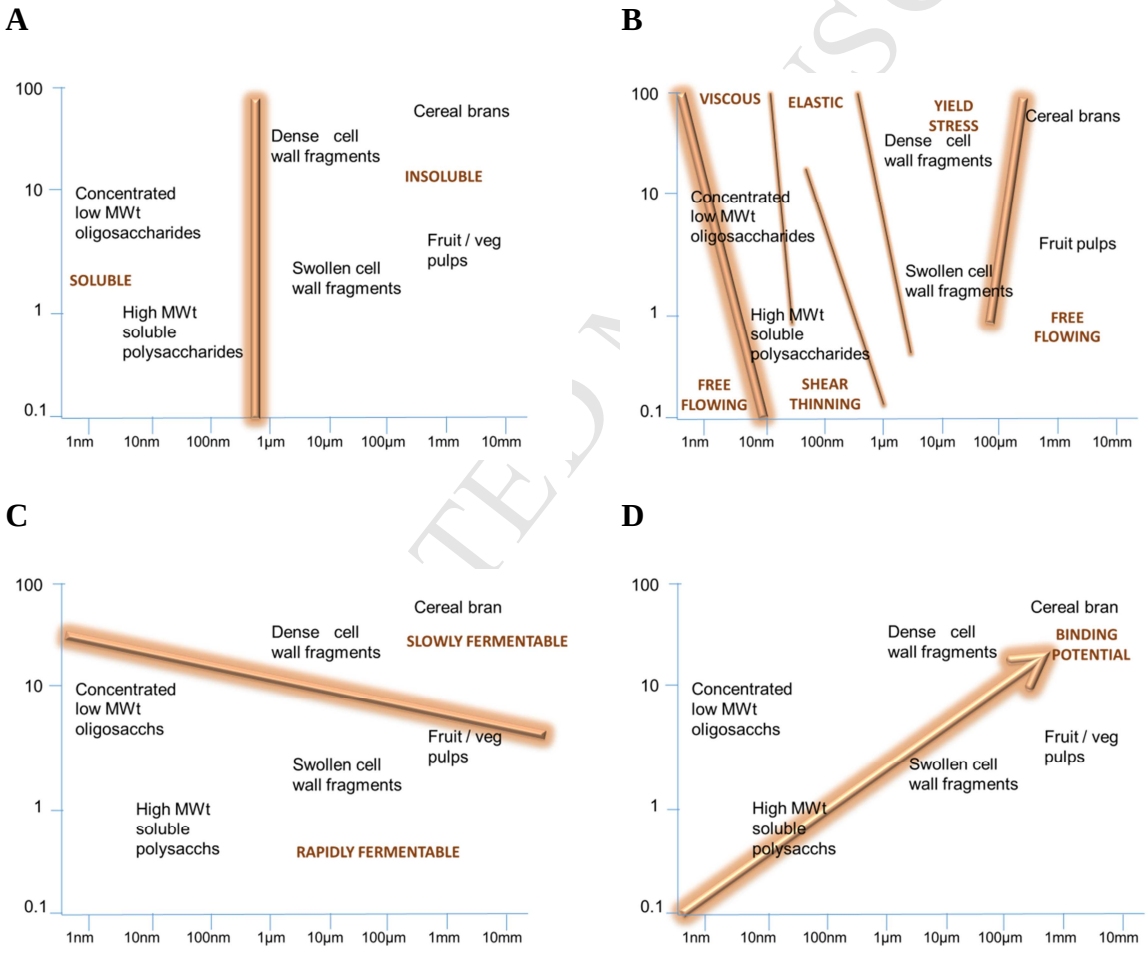

Figure 2. Illustrative expected variations in relevant properties as a function of molecular or particle size (horizontal axes) and concentration or local density (g/100g; vertical axes) in aqueous dietary fibre systems, A. Solubility, B. Flow Behaviour, C. Fermentation rate, D.

Binding potential. The boundary divisions are deliberately broad to emphasise the approximate nature of the size/density cut-offs, and linearity of boundaries is used for convenience.

Flow behaviour of fibre systems can arise from either polymers in solution or swollen particles in suspension, with lower viscosity values (free flowing systems) for either low molecular weight oligosaccharides in dilute solution or suspensions of relatively non-swollen particles that sediment (Figure 2B). At intermediate values of fibre size, the key aspects are elastic response typical of high molecular weight polymers, and yield stress behaviour characteristic of concentrated suspensions. We note that there are of course many other rheological parameters of relevance to the functionality of dietary fibres, with e.g. several types of viscosity (shear, extensional, dynamic). However, each of these can be expected to show systematic responses to fibre systems in different regions of the size/density map.

The fermentation rate of fibre systems is an important parameter because the rate of fermentation is related to the site of fermentation within the large intestine, considering the passage rate. Fermentation is limited under conditions of low water activity as would be found for high concentrations of low molecular weight fibres, but these conditions are not experienced in vivo. Alternatively, fermentation can be slow because the fibre substrate is highly condensed, providing a barrier to efficient utilisation of carbohydrates inside particles of e.g. lignocellulosic brans. As this effect is related to specific surface area, larger particles of less local density should be expected to be fermented at similar rates as smaller particles with higher density. Hence the position of the proposed boundary line in Figure 2C.

Binding of diverse molecules (micronutrients, enzymes, bile salts, mucins etc) to dietary fibres can be driven by chemical specificity or surface interactions, so size and density are not

expected to be the only factors contributing to the extent and/or strength of binding. Nevertheless, where chemical factors have been taken into account, it is expected that molecules/particles of greater local density will provide more efficient binding than less dense systems due to the larger surface energy of the former. Hence the broad directional arrow in Figure 2D. For larger stiffer particles, we also expect surface roughness to have a major influence that can dramatically increase the effective surface area leading to more binding. In contrast, highly hydrated smaller fragments can show low binding due to lower roughness, despite potentially higher nominal specific surface area.

Overall Figure 2 illustrates that a range of features important to dietary fibre functionality have markedly different but systematic behaviours on the size/density plot. This highlights the limited predictive value of categorising fibre as only soluble or insoluble, and suggests that using size/density plots may be a more meaningful way of categorising dietary fibre components such that nutritional functionality can be predicted.

3.3 Challenges and future perspectives

The proposed approach is a broad one, intending to capture all relevant types of dietary fibres. Thus it is necessarily imprecise quantitatively, as generalising behaviour across diverse biological sources and chemical structures would be expected to result in a range of secondary effects on top of those due to size and local density. More quantitative and detailed property maps for individual fibre types at different sizes and densities could in principle be constructed, to compare behaviours between different fibre types. However, the nutritional functionality and preventative health value of dietary fibre is also difficult to quantify precisely, so we expect the maps to be more useful in a semi-quantitative form to compare properties between chemically and biologically diverse dietary fibres.

One challenge in populating the maps will be in quantifying the two coordinates of size and, particularly, local density for individual fibres. We note that the effective size of dissolved oligosaccharides and polysaccharides can be obtained directly from measurements of intrinsic viscosity or from size exclusion chromatography, both of which are related to the hydrodynamic volume. The size distribution of particulate material can in general be readily estimated by microscopy or fractional sieving. Local density is equivalent to concentration for dissolved fibres, but is less easy to determine for particulate fibres. An average density can be obtained from sedimentation volume (or hydration capacity) measurements as long as interstitial volumes are taken into account. The bigger challenge is where there is e.g. an intact cellular structure that has a relatively low average density but is bounded by a thin and dense cell wall which provides an effective barrier (Dhital, Bhattarai, Gorham & Gidley, 2016). Further work is needed to provide realistic local density data for these types of heterogeneous systems.

Once issues of quantification of individual fibre types have been addressed, it will be of interest to consider how best to describe the various regions within the size/density map as a way of communicating the diversity of dietary fibre functionality to consumers. There is a large cohort of consumers who are eager to understand more about why a diet based on plant-based foods, and therefore rich in dietary fibre, is the healthiest option.

A potential use for the proposed maps is to identify whether specific regions of the size/density space are related to individual nutritional benefits of dietary fibre such as cholesterol management, glucose absorption, blood lipid management, fermentation throughout the large intestine, or whether these functionalities overlap in size/density coordinates. If such relationships between size/density co-ordinates and nutritional properties are suggested, then this can form the basis for clinical trials in which e.g. a single fibre source is used with designed differences in size and density. It is possible that a diversity of map

locations for the range of positive nutritional functionalities ascribed to fibre will provide evidence for why a diversity of plant-based foods and therefore fibre types is associated with optimal health outcomes.

One challenge that will need to be addressed is the extent to which individual variation in gastrointestinal physiology and microbiological fermentation over-rides the physical properties of fibres discussed here. A second challenge will be to obtain sufficient data on the physical state of fibres within the digestive tract in humans to understand the mechanisms underlying relationships between ingested fibre size/density and nutritional outcomes. A third challenge is how to simplify the concept for public health messaging, although this needs to be first justified on the basis of property/nutrition correlations and then clinical intervention trials.

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445

Highlights

- Dietary fibre functionality can be related to physicochemical properties
- Bulk structuring, molecular binding and transport barriers all important
- Fibre categorisation on basis of solubility has limited links to functionality
- Molecule/particle size and local density each related to fibre properties
- New categorisation of dietary fibres proposed, based on size/density maps