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Review

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Biosolids-derived fertilisers: A review of challenges and opportunities



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Land application of biosolids is a cost effective way to reuse nutrient in soils.
- Ever changing nature of biosolids contaminants dictates regulatory guidelines.
- Nutrient content in biosolids provides an understanding of baseline agronomic value.
- Extractive technologies can recover and purify valuable constituents from biosolids.
- Prospects for novel granulated fertilisers derived from biosolids are significant.

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ABSTRACT

Soil application of biosolids as an organic fertiliser continues to be a cost-effective way to beneficially utilise its carbon and nutrient contents to maintain soil fertility. However, ongoing concerns over microplastics and persistent organic contaminants means that land-application of biosolids has come under increased scrutiny. To identify a way forward for the ongoing future use of biosolids-derived fertilisers in agriculture, the current work presents a critical review of: (1) contaminants of concern in biosolids and how regulatory approaches can address these to enable on-going beneficial reuse, (2) nutrient contents and bioavailability in biosolids to understand agronomic potential, (3) developments in extractive technologies to preserve and recover nutrients from biosolids before destructive dissipation when the biosolids are thermally processed to deal with persistent contaminants of concern (e.g. microplastics), and (4) use of the recovered nutrients, and the biochar produced by thermal processing, in novel organomineral fertilisers that match specific equipment, crop and soil requirements of broad-acre cropping. Several challenges were identified and recommendations for prioritisation of future research and development are provided to enable safe beneficial reuse of biosolids-derived fertilisers. Opportunities include more efficient technologies to preserve, extract and reuse nutrients from sewage sludge and biosolids, and the production of organomineral fertiliser products with characteristics that enable reliable widespread use across broad-acre agriculture.

Abbreviations: ACT, Australian Capital Territory; ANZBP, Australian and New Zealand Biosolids Partnership; AWA, Australian Water Association; COD, chemical oxygen demand; CSIRO, Commonwealth Scientific and Industrial Research Organisation; EBPR, enhanced biological phosphorus removal; EoWC, End of Waste Code; EPA, Environmental Protection Authorities; EU, European Union; FRV, fertiliser replacement value; NBRP, National Biosolids Research Program; NLBAR, nitrogen limiting application rate; NSW, New South Wales; NT, Northern Territory; OMF, organomineral fertiliser; PFAS, perfluoroalkyl and polyfluoralkyl substances; PFOA, perfluorooctanoic acid; PFOS, perfluororoctane Sulfonate; PRR, partition-release-recover; QLD, Queensland; SA, South Australia; TAS, Tasmania; VIC, Victoria; WA, Western Australia; WWTP, Waste Water Treatment Plants.

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1. Introduction

Biosolids are the main solid end-product of urban wastewater treatment, comprised of sewage sludge treated to achieve a certain quality that reduces or eliminates health and environmental risks and improves beneficial use characteristics. Biosolids production is unavoidable, roughly proportional to population size, and therefore will continue to increase with an increasing global population. In Australia, around 350,000 dry megagrams (Mg) of biosolids were generated in 2021 (Vero, 2022). Biosolids continue to be widely applied as an organic fertiliser and soil conditioner, as this is still the most economical way to beneficially reuse its nutrient and carbon content in soils (Okoffo et al., 2020; Kanteraki et al., 2022). For example, the proportion of biosolids applied to land in the European Union (EU) is about 35 % (Hušek et al., 2022), application of biosolids to arable land is common in the United States (55 %), Canada and New Zealand, (Lu et al., 2012; Raheem et al., 2018), and in Australia, its use in agriculture has increased from 55 % in 2010 to 73 % in 2021, but varies by State and Territory (Vero, 2022).

Biosolids applied to agricultural land can increase crop yield through improvements in soil physico-chemical properties, and agronomic responses are reported to be greater in weathered soils, which are common across Australia (Reid, 2002; Torri and Cabrera, 2017). Beneficial reuse of nutrients from biosolids is important to offset the demand for chemical fertilisers as this can provide a dual benefit, namely: reducing fertiliser price pressures on agriculture, and decreasing greenhouse gas emissions associated with chemical fertiliser production. Since the beginning of 2020, nitrogen (N) fertiliser prices have increased fourfold, while phosphate and potash prices have increased over threefold, and these prices will likely continue to rise (www.cnbc.com, 2022). Between January-December 2021, urea prices increased from AUD256 Mg^{-1} to AUD1026 Mg^{-1} ; mono-ammonium phosphate increased from AUD420 Mg⁻¹ to 952AUD Mg^{-1} and potassium chloride increased from AUD357 Mg^{-1} to AUD822 Mg^{-1} (Austrade, 2022). Prices have continued to rise in January and February 2022 (Austrade, 2022). Additionally, global demand for fertilisers will continue to increase concurrently with increased demand for food, fibre, and biofuels (FAO, 2019). Studies on the emissions avoided by use of nutrients in biosolids have shown that for every Mg of dry biosolids used, around 6 Mg of CO₂ can be avoided (Darvodelsky, 2012). This includes the avoidance of imbedded emissions associated with N fertiliser production via the energy-intensive Haber-Bosch process. There is also global concern over the long-term availability of non-renewable mineral nutrient resources, especially phosphorus (P) (Cordell et al., 2013; Battisti et al., 2022) and potassium (K) (Dawson, 2011; Mehta et al., 2016). It is critical to efficiently recover and safely recycle these nutrients wherever possible to reduce dependency on non-renewable sources, and thereby create sustainable agriculture that also protects the environment (Johnston et al., 2014; Weikard and Seyhan, 2009). This is especially important given that about 80 % of the total phosphate extracted worldwide is used for mineral fertilisers and animal feed additives (Dawson and Hilton, 2011) and >90 % of P ingested by people is excreted and therefore may be recoverable from wastewaters and biosolids into biosolids (Cordell et al., 2009;). Moreover, Australia is a net exporter of food, but is heavily reliant on imported nutrients (Mehta et al., 2016). However, whilst the total nutrient value in biosolids has been widely recognised, limited emphasis has been placed to date on the release and bioavailability of those nutrients, which considerably and directly impacts on the fertiliser replacement value (FRV) of nutrients in biosolids. These aspects are critically evaluated in the current work, to quantify and demonstrate the true agronomic value of biosolids.

The Australian and New Zealand Biosolids Partnership (ANZBP) was established by the Australian Water Association (AWA) to track, promote and support the sustainable management of biosolids in Australia and New Zealand. The ANZBP surveyed community attitudes to the use and management of biosolids in 2010 and 2020 (Jones et al., 2020) with biosolids defined similarly as the treated by-product of wastewater treatment that can be applied to land or used as fuel for power generation. The ANZBP survey results (Jones et al., 2020) showed that the awareness of the term biosolids had increased over time from 33 % in 2010 to 45 % in 2020, and that a large majority of respondents (73 %) were positively disposed toward the use of biosolids for the purposes above. Additionally, over 50 % of community members said that they would be very likely to use biosolids products in their own garden (Jones et al., 2020). The community respondents generally felt comfortable in the knowledge that biosolids use is controlled and regulated (Jones et al., 2020).

In the USA, Canada, and the EU, application of biosolids to land continues to be a dominant practice (Lu et al., 2012), but current legislation imposes varying degrees of restriction to their use in agriculture. In all those countries the most common contaminants emphasized in regulations are heavy metal concentrations in biosolids and soil, and organic contaminants and pathogens in biosolids (Christodoulou and Stamatelatou, 2016). Many differences exist in specific requirements, and, in general, current legislation in the USA focuses on reduction of both pathogens and of pollutants, whereas the EU legislation is directed more toward the regulation of pollutants (Iranpour et al., 2003). The processes involved in the development of

the regulatory framework in Australia were based on, or modified from European and North American guidelines (Hill, 2005). However, the soils, climatic conditions, and agricultural practices in Australia are sufficiently different to those in other parts of the world to justify use of locally developed guidelines in Australia to protect the environment and the human food chain (Whatmuff and Osborne, 1992; McLaughlin et al., 2000; Hill, 2005). There are several features of soils in Australia that distinguish them from soils commonly found in Europe and North America: specifically, many of the soils of Australia are pedologically very old and highly weathered, have a very low level of soil fertility and consist of variable charge minerals. The above differences are important when considering the reactions of metals in soil (McLaughlin et al., 2000). Heavy metals are one key contaminant class addressed by regulatory frameworks, which is important for agricultural use of biosolids. However, the impact of regulations on source control and heavy metals in Australian biosolids has not been previously investigated, and this is illustrated in the current work by tracking heavy metals in biosolids by consolidating and presenting literature data published over several years. In addition, the review considers the concept of bioavailability of heavy metals and the impact that this may have on the risks associated with the agricultural use of biosolidsderived fertilisers.

Typically, biosolids have been applied to land in their original form. Sustainable farming practice aims to balance nutrient inputs with outputs. An 'ideal' fertiliser supplies nutrients only on the basis of plant demand, in relation to its phenological stage (Trinchera et al., 2011), and to prevent environmental impacts associated with over-application or poorly timed application (Sakrabani, 2020). There are significant differences in nutrient availability between biosolids and mineral fertilisers. Nutrients in mineral fertilisers are generally in a soluble form, and when applied are immediately plant-available. In contrast, a large proportion of nutrients in biosolids are in organic forms which must first to be mineralised to be plantavailable. This presents both a challenge and an opportunity by blending or reacting mineral fertilisers with organic fertilisers to produce so-called organo-mineral fertilisers (OMF). Such fertiliser mixes or products could balance the rapid release properties of nutrients from mineral fertilisers with sustained slow mineralisation and release of nutrients from an organic component (e.g. composted manure, Abbott et al., 2018), and, additionally, could improve soil structure, drainage, water availability, and increase soil carbon to promote crop growth (Sakrabani, 2020; Antille et al., 2013b). The use of such products represents a technological advancement but requires consideration of several important product and application-related factors to enable successful use in broad-acre agriculture. To date, there has not been a review and evaluation of these important considerations, and this is addressed in the current paper.

Despite the benefits to date of regulations to ensure that stability classes are met, vector attraction potential is minimised, and contaminants are restricted to below levels where significant environmental impacts would be expected, challenging contaminants in biosolids continue to emerge and these are threatening the long-term viability of the direct agricultural use of biosolids. Regulatory approaches have attempted to engage with such emerging contaminants, but microplastics and nano-plastics have become a significant concern for environmental and human health due to their ubiquitousness and because of new research suggesting links to significant adverse impacts (Section 3). Consequently, there have been indications that thermal processing of biosolids (e.g. gasification, pyrolysis or incineration) may be a convenient way forward to destroy persistent contaminants of concern (Hušek et al., 2022). For this reason, there has been increased research interest in technologies to extract valuables (N, P, K, and others) from biosolids or its precursor sewage sludge (Gianico et al., 2021) before the biosolids or sludge is sent to thermal process. To date, there has not been a targeted review on such extraction technologies specifically in support of recovery and preserving of nutrients before thermal processing of biosolids to use in balanced organic fertiliser formulations; as such, these aspects are addressed in the current work.

The current review was conducted to promote the safe beneficial reuse of organic fertilisers derived from biosolids in agriculture. This is done by providing, for the first time, a critical evaluation of the above-named important aspects of agronomic value and bioavailability of nutrients in biosolids, regulatory influences and opportunities for benefits in contaminant levels due to tighter catchment control, targeted extractive technologies to recover and preserve nutrients before thermal processing of biosolids, and using these nutrients together with biochar from thermal processing to formulate balanced OMF suitable for broad-acre agriculture.

2. Agronomic value of biosolids constituents

Application of biosolids for agriculture has significant potential because of the volumes produced that could be used for its nutrients and carbon content (fertilizing and- soil conditioning), at comparatively lower costs. A substantial body of past research in Australia has focussed on biosolids effects on soils and plants (Table 1), including important work dating as far back as the 1980–1990s (De Vries, 1983; Jakobsen and Willett, 1986; Dann et al., 1989; Barry et al., 1998). Included in this has been the Australian National Biosolids Research Program (NBRP) established in 2002 by the Australian Commonwealth Scientific and Industrial Research Organisation (CSIRO). The NBRP conducted national trials of biosolids under a wide range of conditions, including various soil types, climates, and cropping systems, to evaluate the true agronomic benefits of biosolids in agriculture, and to understand how well overseas research findings might translate into the Australian context. For this, a total of 17 field sites were established in five Australian States and examined both potential beneficial and

Table 1

Physico-chemical and biological responses of soil amended with biosolids. Outcomes of Australia based published scientific experiments.

Soil property	Effect	References
Aggregate stability Bulk density CEC EC	No effect No effect Increased Increased	Ives, 2012 Ives, 2012 Munn et al., 2001; Sarooshi et al., 2002; Sarooshi et al., 2002; Rajendram et al.,
Heavy metals	Increased, but below regulatory threshold levels	2011; Ives, 2012; Antille et al., 2020 Joshua et al., 1998; Dumbrell, 2005; Munn et al., 2001; Whatmuff, 2002; Cooper, 2005b; Bell et al., 2006; Pritchard and Collins, 2006; Warne et al., 2008; Eldridge et al., 2009; Nash et al., 2011; Rajendram et al., 2011; Qi et al., 2011;
Infiltration/Surface runoff	Decreased	Joshua et al., 1998
Macronutrients: N, P, K, S	Increased	Munn et al., 2001; Pu et al., 2004; Stokes et al., 2004; Cooper, 2005a; Pritchard and Collins, 2006; Pu et al., 2008; Beshah, 2010; Nash et al., 2011; Rajendram et al., 2011; Ives, 2012; Pu et al., 2012; Albuquerque, 2018; Antille et al., 2020; Rahman et al., 2021
Microbial biomass	Increased	Madejón et al., 2010; Ives, 2012; Thangarajan et al., 2015; Chowdhury et al., 2016; Wijesekara et al., 2017.
рН	Decreased	Rajendram et al., 2011; Nash et al., 2011
	Increased	Munn et al., 2001; Sarooshi et al., 2002; Cooper, 2005a; Murtaza et al., 2011; Rahman et al., 2021
Total or organic C	No effect Increased	Pu et al., 2004; Ives, 2012 Munn et al., 2001; Sarooshi et al., 2002; Pritchard and Collins, 2006; Nash et al., 2011; Bolan et al., 2013; Wijesekara et al., 2017; Albuquerque, 2018
Water holding capacity	Increased	Belyaeva et al., 2012
Yields	Increased	Pu et al., 2004; Stokes et al., 2004; Cooper, 2005a; Corrêa et al., 2005; Pritchard, 2005; Warne et al., 2008; Beshah, 2010; Madejón et al., 2010; Lamb et al., 2012; Murtaza et al., 2012; Bolan et al., 2013; Antille et al., 2020

deleterious effects. Across all NBRP sites, biosolids were applied to supply nutrients at similar levels to historic fertiliser use. In general, biosolids were applied according to States and Territories guidelines that specify the N-limiting biosolids application rate (NLBAR) and was found to deliver sufficient nutrients for at least 1–2 annual cropping cycles (averaged across all sites in the NBRP) without needing mineral fertiliser (McLaughlin et al., 2008). Moreover, the application of biosolids was observed to have a positive effect on crop yields and plant nutrient contents with the main benefits probably being due to N and P addition with biosolids (Stokes and Surapaneni, 2004; McLaughlin et al., 2008). The NBRP continues to be the most comprehensive and significant program of research to date concerning biosolids use in Australian agriculture.

Typically, biosolids contain high concentrations of N, P, K, and sulphur (S), and several micronutrients, including copper (Cu), zinc (Zn), calcium (Ca), magnesium, boron (Bo), molybdenum (Mo), and manganese (Mn). The application rate of biosolids in Australia is determined by total N and NLBAR. However, one concern with NLBAR is typical low N:P ratios in biosolids in terms of crop fertiliser requirements, so that biosolids application based on N could result in excess P. When biosolids are routinely applied, this can result in progressive build-up in soil P levels, increasing the risk of P transport to water courses by erosion and runoff (Warne et al., 2008; Pritchard et al., 2010). To demonstrate variability of nutrients content in biosolids across Australia, data from over 70 relevant peer-reviewed articles, technical reports and PhD studies were compiled for the present review (Supplementary Materials, Tables S1, S4, and S5). Biosolids properties and composition was observed to vary between analysis batches and between wastewater treatment plants (WWTPs). This was plausibly due to differences in the specific treatment processes in place and wastewater conditions changing with time or being different across locations (McLaughlin et al., 2000; Ukwatta and Mohajerani, 2015).

The agronomic value of biosolids can be estimated based on total nutrient content and per unit nutrient price. For the current work and using the average quantities of biosolids applied to agriculture in Australia over 2010–2021 and N, P and K contents in Australian biosolids (Table S1), the total maximum nutrient value was estimated could be up to 33 million AUD per year for nominal prices of AUD2.23 kg⁻¹ N; AUD3.63 kg⁻¹ P, and AUD1.37 kg⁻¹ K (Supplementary Material, Table S2).

To determine true FRV, it is important to consider nutrient plantavailability (Warne et al., 2008), which for biosolids can vary considerably (Table 2). Nutrients in biosolids are typically slow-release, for example, with 15-50 % of the N and P becoming available within the first year and an additional proportion in subsequent years (McLaughlin et al., 2008; Pritchard et al., 2010). The slow mineralisation of biosolids can help maintain plant-available N during periods of rainfall when conventional fertiliser is at significant risk of leaching from the crop root zone (Pampana et al., 2021). The efficiency of N input from industrial fertiliser to agriculture is also generally poor, with an estimated 40-70 % N typically dissipated into the environment (Chojnacka et al., 2022). Guidelines in Australia define N mineralisation rates for biosolids applied to land over one year of application at 15 % for anaerobically digested biosolids and 25 % for aerobically digested biosolids. However, a study conducted by Eldridge et al. (2008) in NSW found that up to 50 % of total N in land applied biosolids could mineralise in the first 2 months after application. In TAS, Ives et al., 2010 observed that 35 % of total N in anaerobically digested biosolids being land applied could mineralise in 59 days. In warm, subtropical QLD, mineralisation rates also averaged 55-60 % of the applied organic N in the first

Table 2

Availability^a of N and P in biosolids applied to land across Australia. Data from multiple sources with references given in the Supplementary Material, Table S4.

Nutrient	Availabili	Number of				
	Min	Max	Mean	Std. Dev.	Median	data points
Ν	0.01 %	38.7 %	11.2 %	9.3 %	9.4 %	52
Р	0.37 %	35.0 %	13.5 %	11.4 %	10.8 %	10

^a Calcium chloride extractable.

6–9 months following biosolids application, with at least 30 % (and in some cases 60 %) of the applied organic N being mineralised within the first 6–8 weeks after incorporation (McLaughlin et al., 2007). These results indicate a need to understand nutrient release characteristics to better understand the true FRV of biosolids, including globally.

Other macro and micro nutrients in biosolids (e.g., S, Ca, magnesium, Fe, Mn, Cu, Zn, B, Cl and Mo) may also become increasingly valued into the future, together with the potential soil amelioration effects of biosolids applied to degraded soils (e.g. lacking soil carbon, or with poor moisture holding capacity) and/or nutrient-depleted soils. These benefits are not currently well quantified and requires further research investigation, including under relevant field conditions, and climate context. In general, it is expected that multiple benefits of biosolids will gain in importance over time, as will the perceived value of biosolids as organic fertiliser, especially as these factors become better understood.

3. Environmental and health concerns with biosolids use in agriculture, current regulatory controls

Production and application of biosolids in Australia is regulated by State-based Environmental Protection Authorities (EPA) or equivalent bodies, according to guidelines specific to each State or Territory (Supplementary Material, Table S3). These guidelines set out quality assurance requirements and best management practices for reuse, and gain legal standing where they may be called up in relevant legislation or referred to in an environmental license of a facility. New South Wales was the first state in Australia to develop guidelines for the beneficial reuse of biosolids (New South Wales Environmental Protection Authority, 1997), and the subsequent development of guidelines in other states and the national biosolids guidelines that followed, largely mirrored the NSW guidelines (McLaughlin et al., 2007). The basic structure of all the guidelines is similar, comprising contaminant grading; stabilization grading (i.e., pathogen and vector attraction, odour potential); and management controls, including sampling and monitoring. An overall combined quality grade (stability and contaminant) determines permissible uses for the classified biosolids. The Australian approach to managing contaminants in biosolids has been relatively similar to that used in the USA, except for the actual limits imposed. Australian guidelines have been suggested to include significantly stricter requirements than that required in the USA (Reid, 2002). Also, to ensure that excessive levels of contaminants are not added to otherwise clean soils, guidelines specify that both biosolids and the soils to which they are to be applied are to be monitored for the levels of relevant contaminants. The guidelines also include several further controls relating to soil pH, soil slope, soil water regime, and proximity to watercourses, roads, property boundaries, and sensitive receptors (e.g. residences), as these influence the risk of adverse environmental and amenity impacts. To demonstrate the level of biosolids-specific guidance provided, the example in the Australian State of Queensland (2020) is where an End of Waste Code (EoWC) for Biosolids deals with various controls and factors and in fact includes trigger values in soils for perfluoroalkyl and polyfluoroalkyl substances (PFAS) (Hall et al., 2021) (see further below).

Unfortunately, organic contaminants are common in industrial, medical, and household products and applications and therefore usually end up in human-derived wastewater (Kinney et al., 2006), many of which are persistent and potentially bioaccumulative. Organic contaminants can include pharmaceuticals, hormones, detergent metabolites, fragrances, plasticizers, and pesticides. Organic pollutants of primary interest include organochlorine pesticides, polychlorinated byphenyls and polychlorinated dioxins/furans. Importantly, most organic chemicals are present in biosolids in Australia at low concentrations (Smith, 2009) (often below detection limit) and have therefore been suggested as unlikely to pose an issue for land application of biosolids to soils (Clarke et al., 2008; Clarke et al., 2010). However, the nature and types of pollutants found in biosolids are constantly evolving with improvements in measurement and identification capabilities, and this is an on-going global challenge for policy to manage 'unknown' or 'emerging' contaminants. Three options to do this have been previously suggested (Clarke, 2014): (1) regular national biosolids surveys for emerging pollutants; (2) development of an Unregulated Contaminant Monitoring Regulation program; and (3) development and application of biological-based assays for generalised toxicity that can be related to relevant human/ecological receptors.

One emerging area of research interest is the link between effects of land-applied materials such as biosolids, on soil microbial health. Moderate applications of biosolids have been suggested could increase the diversity of the soil ecosystem, as the additional organic matter and nutrient inputs support the growth of microbial populations, leading to an increase in diversity (Goyal et al., 2008). However, the observed impact of biosolids on soil microbial diversity may not always been positive (Markowicz et al., 2021; Goyal et al., 2008). For example, the field application study of Mossa et al. (2017) using biosolids indicated that for soil samples collected from 17 maize fields, soil microbial diversity decreased with increasing zinc (Zn) concentrations in soils. This suggests that above a certain level, heavy metals accumulation of biosolids might offset the positive impact of organic matter on soil microorganisms. Currently, regulatory approaches do not address soil microbial ecotoxicity effects, whereas into the future the further development and use of ecotoxicity tests may become more prominent in combination with traditional chemical analyses. In the future this may enable the evaluation of the potential effect of toxic substances (including those in amendments such as biosolids) on soil microorganisms (Giannakis et al., 2021). However, soil microbial ecotoxicological data for the effect of pollutants from biosolids on Australian soils and organisms still are sparce (Broos et al., 2007).

One group of emerging contaminants which has received increasing attention is PFAS and Perfluororooctane Sulfonate (PFOS). PFAS substances are used in a wide variety of applications in industry and household products. The ANZBP conducted a national survey on the presence of PFAS in biosolids in which major utilities voluntarily shared data for over 100 samples from 13 different sewage treatment plants around Australia (Hopewell and Darvodelsky, 2017). This data found that PFAS concentrations were well below proposed biosolids limits, and therefore may be posing a low level of risk when land applied. Similarly, the average level of PFOS measured in biosolids was around 0.5 % of the suggested safe level for agricultural use, and the maximum level of PFOS measured at all sites was also lower than the suggested safe level by a factor of about 11, including for two sites with a known history of elevated PFOS (Darvodelsky and Hopewell, 2018). The ANZBP investigation hence concluded that: a) PFOS and PFAS were present in biosolids at detectable levels; b) average values of PFOS measured in Australian biosolids were around 7 % of the calculated Health Investigation Level; and c) levels of Perfluorooctanoic acid (PFOA) detected were significantly lower than Health Investigation Levels suggested by the Australian Government (Hopewell and Darvodelsky, 2017). Accordingly, it was recommended that limits in biosolids be adopted and be routinely reviewed as further data became available (Hopewell and Darvodelsky, 2017).

Heavy metals in soil and their transfer to the food chain has long been a key consideration for land application of biosolids and biosolids-derived fertiliser products (Hušek et al., 2022). For this reason, a significant amount of data was found in the literature for the heavy metal content of biosolids across Australia (Supplementary materials, Table S6). These data were collated and assessed for the current work to determine whether progress in regulatory controls of heavy metals, such as via pressure on industrial catchments to reduce heavy metals discharged to sewer, could have led to changes in heavy metals in biosolids over time. Encouragingly, total heavy metal content in biosolids were observed to reduce over time (Fig. 1). This could have been caused by improved technologies/products/practices, and due to more stringent source control for domestic WWTP catchment (e.g., industrial flows), and shows a potential for developments to positively influence biosolids quality over time.

Most biosolids regulations around the world have thresholds for total heavy metal concentrations in biosolids (being the readily measurable quantity) and some in Australia (e.g. EoWC in Queensland) also include adjustments for background soil heavy metal levels. During the NBRP research study, critical soil concentrations of Cu and Zn were assessed in terms of adverse effects on microbial processes and plant productivity, and found to be affected by soil pH, clay content, organic carbon content and cation exchange capacity (Warne et al., 2008). This led to a set of soil-specific threshold limits being proposed for Cu and Zn. The properties of biosolids also play a crucial role in determining the mobility of heavy metals in soil (Merrington et al., 2003; Haynes et al., 2009) and heavy metal mobility can be highly variable. This was demonstrated by data from our own work and that of Oliver et al., 2005, (Table 3) and indicated that heavy metal mobility is ideally assessed on a case-by-case basis to determine the potential for detrimental environmental impacts and benefits (Oliver et al., 2005). Although it is understood that the available concentration controls the contaminant toxicity, this issue remains a key knowledge gap, and accordingly biosolids guidelines may be imposing excessively conservative threshold levels if based on total heavy metals.

Microplastics and nano-plastics are an emerging contaminant in biosolids of major concern to the safe direct beneficial reuse of biosolids in agriculture. Both microplastics and nanoplastics are small plastic particles, with respective diameters of 1-5000 µm and 1-1000 nm, and are now ubiquitous in the environment (Leusch and Ziajahromi, 2021). Plastics in domestic wastewater, which end up in biosolids, originate from plastic-containing household products via normal household cleaning and washing (e.g. synthetic fibers from clothes washing), plastics from cosmetics and personal care products, and abrasive plastics in cleaning agents (Okoffo et al., 2020). An investigation of microplastics in Australian biosolids sampled 82 WWTPs across Australia and detected plastics concentrations ranging from 0.1 to 9.6 mg. g^{-1} dry weight, with polyethylene being the predominant plastic detected (Okoffo et al., 2020). When biosolids are land applied in agriculture, microplastics become part of the soil and can influence its properties, and can be bioavailable to animals and plants, or can migrate into aquatic ecosystems (Hušek et al., 2022). Microplastics can be hazardous and elicit chemical, physical, and biochemical toxicity (Bläsing and Amelung, 2018; Okoffo et al., 2020). Currently, there is unfortunately a lack of standardized and applicable methods to identify and quantify microplastics in complex samples such as wastewater and biosolids, and this has increased uncertainty in microplastics assessments (Ziajahromi et al., 2017). However, despite this, it will be challenging to completely eliminate microplastics from domestic wastewater and biosolids, and accordingly, some have suggested the direct application to agriculture may not be a viable future option (Hušek et al., 2022). Instead, thermal processing of biosolids (e.g. pyrolysis, gasification) has been proposed for the full destruction of plastics and other persistent contaminants (Hušek et al., 2022).

4. Extractive technologies for preparation of biosolids-derived fertilisers

Unfortunately, thermal processes are destructive to soil-active carbon compounds (e.g. humic substances) and N (see Section 4.1) and can be detrimental to the bioavailability and purity of P (see Section 4.2). Accordingly, there has been increasing interest in technologies for upstream extraction of nutrients and carbon compounds before the biosolids are sent to thermal processing. These can then be safely beneficially reused (Gianico et al., 2021). These technologies are reviewed in this section.

4.1. Nitrogen recovery

Into the future, when WWTPs are converted into resource recovery facilities, the destructive dissipation of carbon and N via the activated sludge process will likely be replaced with energy-efficient extractive technologies to recover N and carbon in useful forms. In this regard, a useful partition–release–recover (PRR) framework has been previously described for the efficient selection and integration of such technologies (Batstone et al., 2015), whereby; (1) N is partitioned from the main water line of the WWTP to the sludge line via bioassimilation; (2) N in the sludge is then released via digestion (ideally anaerobic with simultaneous energy recovery) and; (3) is finally N is recovered in forms suitable for the intended



Fig. 1. Total heavy metal content in Australian biosolids over the period 1970–2022, mg/kg dry solids. Data were collated from a large range of literature sources as cited in the Supplementary Material, Table S5.

end-use (e.g., fertiliser). For the partition step (step 1), anaerobic photoheterotrophic mediators have been of particular interest, efficiently using light energy to assimilate carbon and nutrients from wastewater into a protein-rich microbial biomass (Capson-Tojo et al., 2020). Limited investigations using pot trials have already shown that such microbial biomass almost performed as well as chemical fertilisers as a nutrient source for pasture grass (Zarezadeh et al., 2019). For the release step (step 2

above), soluble products from a first-stage fermentation at lower pH and short hydraulic retention time may be useful as the biodegradable carbon for bio-assimilation in the partition step (Batstone et al., 2015). A recent novel process also explored the direct enhanced recovery of up to 50 % ammonia from sludge fermentation under vacuum (Okoye et al., 2022). Such developments will be important to reduce the energy consumption for N recovery so that it at least becomes comparable with that used for N

Table 3

Percentage of bioavailable ^a to total c	concentration of sele	ected heavy metals	in Australian	biosolids.
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*				•			
Biosolids location	Cd	Pb	Cr	Ni	Cu	Zn	References
Site A	0.84	0.00	0.06	6.58	0.53	0.66	Unpublished data, Waste to Profit project, Ramirez et al., 2021
Site B	1.33	0.00	0.12	2.52	0.87	0.07	Unpublished data, Waste to Profit project, Ramirez et al., 2021
Werribee	6.90	n.a. ^b	n.a. ^b	8.70	0.50	8.30	Oliver et al., 2005
Chelsea	0.20	n.a. ^b	n.a. ^b	3.40	0.60	0.70	Oliver et al., 2005
Bolivar	1.60	n.a. ^b	n.a. ^b	5.90	4.70	0.40	Oliver et al., 2005

^a Calcium chloride extractable.

^b Not reported.

manufacturing as chemical fertiliser (Batstone et al., 2015). This remains an important aspect for future research and development.

4.2. Phosphorus recovery

Globally, it has been estimated that 20 % of mineral P consumed is excreted by humans and thus potentially recoverable (Cordell et al., 2009; Batstone et al., 2015), whilst about 80 % of the total non-renewable rock phosphate extracted worldwide is used for mineral fertilisers and animal feed additives (Dawson and Hilton, 2011). With a likely increase in thermal processing of biosolids into the future, the impact of such processing on plant availability of P would be important. For example, the study of Mackay et al. (2017) showed that extractable P in by-products of biosolids converted via four thermal conversion processes (pyrolysis, incineration, and two forms of gasification) was lower than in unprocessed biosolids. Moreover, it has been suggested (Mehta et al., 2015) there could be a competition with incineration between operating at low temperatures (<700 °C) to ensure a high fertiliser efficiency of P (Thygesen et al., 2011) vs. operating at higher temperatures >900 °C to minimise nitrous oxide emissions (Gutierrez et al., 2005). For these reasons, it is important to target the upstream recovery of P via wastewater treatment or from sewage sludge before biosolids is sent to thermal processing. Phosphorus can be recovered from the sludge line of a WWTP via minerals precipitation, albeit at high and somewhat limiting operational cost (Raheem et al., 2018). However, the recovery of P can be facilitated by accumulation of P into biomass via enhanced biological P removal (EBPR), and with a subsequent release step (e.g. thermochemical or biochemical), this P can be solubilised to be more efficiently recovered at higher concentration via mineral precipitation (e.g. struvite) (Yuan et al., 2012). Struvite technology is already commercially available, producing struvite fertiliser with favourable characteristics and generally low levels of contamination (Muys et al., 2021).

4.3. Humic substances

There has been significant interest in extraction/recovery of molecules from sewage sludge with an organic soil amendment benefit (Núñez et al., 2022). Humic substances is one such class of molecules found in sludge, said to make up an estimated 10–15 % of the total sludge dissolved organic matter (Li et al., 2013; Xiao et al., 2020), and comprised of humic acids and fulvic acids (Xiao et al., 2020). The benefits of humic substances as biostimulants of plant growth have been well-documented (Jindo et al., 2020). Humic substances are inherent components in sludge produced via biological release during anaerobic digestion and are also produced and/ or chemically altered via thermal or chemical pre-treatment of sludge (Xiao et al., 2020). Humic acid is said to be recoverable from sludge in an up-concentrated liquid form via membrane filtration (Núñez et al., 2022).

4.4. Section summary

Overall, to recover nutrients and organic amendment compounds from wastewater and sludge before biosolids is thermally processed, various upstream recovery technologies would need to be integrated into or replace conventional WWTP processes that currently destructively dissipate carbon and N (Fig. 2). Several PRR technologies are commercially available and have been reasonably widely applied as individual technologies. However, their successful integration into a whole of plant context at full-scale remains a development gap (Batstone et al., 2015). Following the description of technologies above, there is an opportunity to extract/recover P and N into chemical fertiliser forms which are readily bioavailable, and these



Fig. 2. Schematic overview of an alternative future scenario where conventional WWTPs are converted into closed-loop resource recovery centres, in this case to produce balanced biofertilisers for broad-acre cropping applications.

can then be used in novel OMF formulations with properties that target broad-acre cropping applications.

5. Biosolids-derived fertiliser products tailored for broad-acre farming

Key drivers for organic-based fertilisers in agriculture are (1) technological developments that are enabling the production of high-quality products, (2) improvements in application techniques for field spreading, cost advantages compared with mineral or synthetic fertilisers, and (3) the need to maintain soil carbon and fertility levels thus allowing for increased circularity of carbon and nutrients (Chambers et al., 2003; McCabe et al., 2020; Burggraaf et al., 2020). The following section reviews key considerations associated with biosolids-derived fertilisers in agriculture, to facilitate their widespread adoption and use for broad-acre crop production (Antille et al., 2013b; Antille et al., 2017).

5.1. Physical and mechanical properties

For solid fertiliser products, the physical (e.g., density properties, particle size and size distribution) and mechanical (e.g., static particle strength) properties are very important to enable successful handling, transport, storage and mechanized application. For example, materials that exhibit a moderately high crushing strength are also able to resist forces imposed by handling, storage, and spreading without significant shattering, dust formation, or caking. A breaking force of 15 Newton has been suggested as a lower limit to avoid particle fracture during handling and field spreading (Hignett, 1985). Unlike fertiliser spreading equipment, machinery used for application of solid organic materials (and likely biosolids-derived organic fertilisers) does not necessarily allow for high degree of control over the placement and uniformity of the material being applied. Consequently, the distribution of the material on the ground can be less uniform than conventional (granular or liquids) fertilisers, both along the direction of travel and across the working width of the machine. The application rates are controlled by the forward speed and the calibration of the metering system, and the physical properties of the material being applied are important for a single application (e.g., changes in moisture content, density, particle size). Dimensional analyses (e.g., Gregory and Fedler, 1987) have shown that granular materials flow, such as during discharge from a fertiliser spreader or during loading/unloading operations, depends on density properties. Density properties are also related to the volume needed for storage and transport, and together with particle size and size distribution are important for field-spreading equipment (Antille et al., 2015). For example, the uniformity of distribution of fertiliser materials during field spreading is influenced by particle size and size distribution and particle density, because these properties influence particle segregation and aerodynamics (Hofstee and Huisman, 1990; Bradley and Farnish, 2005). This is important because uneven spreading increases the risk of nutrient losses to the environment and can influence nutrient use efficiency (Jensen and Pesek, 1962). For this, a coefficient of variation of about 10-12 % in particle size has been suggested to be a threshold above which a loss in yield and potentially reduced quality in grain in terms of protein content could impose financial penalties (Miller et al., 2009). Studies by Antille et al. (2013a, 2015) showed that the optimum particle size range of granular biosolids and biosolids-derived fertilisers were between 1.10 and 5.50 mm in diameter for conventional twin-disc spreaders.

5.2. Chemical composition

Biosolids generally have low N:P ratios (Section 2). Moreover, biosolids are generally well-supplied with P (range: 5-12 % total P as P_2O_5) but have less K (typically, <2 % total K as K_2O) (Krogmann and Chiang, 2002). This is important because land application based on crop N or K requirements then risks the progressive build-up of soil P levels. While high soil P levels do not necessarily result in plant toxicity, elevated P increases environmental risk associated with potential soil P transport to surface water and groundwater. Blending with mineral or synthetic fertilisers can be used to correct for imbalance and/or inconsistent chemical composition (nutrients and C:N ratio) between different batches (Sommers, 1977) to achieve a desirable N:P:K ratio. This may be required to suit specific soil and crop requirements. By optimizing the nutrient composition and adjusting the nutrient application rate, the nutrient recovery in the crop and therefore use-efficiency can be improved (Antille et al., 2013c; Antille et al., 2017). Potential build-up of heavy metals in soil and uptake by crops grown on the soil can lead to their subsequent transfer to the food chain (Jones and Johnston, 1989; Jones, 1991). The associated risks should be appropriately managed, also by considering background soil heavy metal levels and heavy metal mobility (see Section 3).

5.3. Agronomic efficacy

Information available in the scientific literature suggests that FRV of organic materials, including biosolids and biosolids-derived fertilisers, can often be <40-60 % of that using urea or ammonium nitrate (e.g., Lalor et al., 2011; Petersen, 2003; Ashekuzzaman et al., 2021). However, some nutrients present in organic materials are in organic forms and therefore could undergo delayed or slow mineralisation to become plant-available. Research to date has been particularly interested in understanding the mineralisation-release characteristics, and how the application of straight fertilisers and organic sources (e.g. composted manures) in splits could be used to increase the overall nutrient recovery in crops and mitigate potential yield penalties by inadequate nutrient supply with organic materials alone. The correct synchronization of nutrient supply (from the soil/organic material) with demand (from the plant) is a key factor influencing nutrient use efficiency. Knowledge of the soil/crop/environment specific factors governing nutrient transformations in soil and the ability to predict such processes is a key requirement for improved nutrient use efficiency of organic materials, and for timely field application.

Agronomic performance may also be improved by blending with mineral fertilisers until the desired nutrient ratio and appropriate mineralisation-nutrient release characteristics can be obtained. This has included the reactive conversion of the blend to a compound referred to as an OMF. The product specifications for OMF by coating biosolids granules with mineral sources of N (as urea) and K (as potash) was reported in a series of studies by Antille et al. (2013b, 2015), as shown in Fig. 3), with N:P: K ratios of approximately 10:5:5 and 15:5:5. The main advantage of OMF vs. conventional blending is that the physical and mechanical properties of OMF particles can be made more consistent, which can provide greater certainty with handling and field application. For blended materials, if the physical and mechanical properties of the constituents in the blend are significantly different, segregation can occur (Bridle et al., 2004), which could affect aerodynamic behaviour and uniformity of distribution during field spreading (Grift et al., 1997). Moreover, for OMF, the mineral fraction represents a source of nutrients that are rapidly released and are readily available for crop uptake, whilst the organic fraction undergoes slower mineralisation to provide more sustained nutrient supply following soil application (Smith et al., 2020). Unfortunately, the rate of nutrient release from the organic fraction of OMF can be difficult to predict and this will be important into the future to manage benefits and impacts from agronomic and environmental perspectives (Antille et al., 2014a, 2014b). The conversion of biosolids into OMF products tailored to meet specific soil and crop needs represents a technological advancement compared with ways that biosolids have been traditionally used in agriculture.

5.4. Biochar as a co-component of OMF

Incineration of sewage sludge is still a common practise across Europe since restrictions to landfilling of sewage sludge were introduced with the EU Landfill Directive (99/31/EC) (CEC (Council of the European Commission), 1999). However, due to a high cost and poor public perception of sludge incineration (Raheem et al., 2018) pyrolysis and gasification have attracted increasing interest as well-known thermal processing alternatives. Pyrolysis and gasification produce biochar as a co-product (Raheem et al.,



Fig. 3. OMF granules and biosolids, after Antille et al., 2013b, with permission.

2018). The use of biochar in soils and agriculture has attracted considerable research interest in recent years (Abbott et al., 2018), because due to its unique physicochemical features, biochar from sewage sludge or biosolids has the potential to be utilised as a soil amendment fertiliser (Lehmann and Joseph, 2015). For example, depending on its characteristics, biochar could increase soil structure, water retention capacity and nutrient retention as a soil conditioner, and as a fertiliser it could deliver nutrients to plants, increase microbial activity and reduce nutrient losses due to leaching and volatilisation (Cayuela et al., 2013; Kloss et al., 2012). Biochar can also have sorption properties that mitigate N leaching, and influence relevant soil microbial processes to reduce N losses in some soils (Shanmugam et al., 2021). Lastly, biochar has also previously been suggested as a means to sequester carbon in soils (Marris, 2006).

Thermal processing results in mass destruction/volume reduction/upconcentration of contaminants in ash or biochar. Some studies have been concerned that use of biochar from biosolids may increase the heavy metal accumulation in the soil-plant system, posing a potential threat to agricultural soil (Song et al., 2014; Yue et al., 2017). However, several studies have demonstrated that heavy metals can be immobilised in biochar derived from biosolids, reducing their bioavailability and reducing the risk of soil-plant contamination (Hossain et al., 2010; Faria et al., 2017; Sousa and Figueiredo, 2015); albeit that it is important to note that heavy metals may be immobilised to varied extents depending on the pyrolysis conditions, the resulting biochar characteristics, and soil and crop effects (Jin et al., 2016; Patel et al., 2020). Considering that biochar would likely be available as a by-product from end-of-pipe thermal processing of biosolids, the interest in inclusion of biochar in soil amendments or as fertiliser cocomponent will likely increase over time. There has already been a move to include biochar as a co-ingredient in amendments, including with conventional fertilisers (Abbott et al., 2018).

Other than understanding the agronomic benefits of using biochar, future research will need to identify preferred pyrolysis conditions that produce biochars with the desired physical structure and composition for soil amendments (Abbott et al., 2018). This can then develop targeted and sustainable fertiliser formulations to support soil health and provide plant benefits (e.g., Yeboah et al., 2017).

6. Opportunities, and recommendations for further work

There has been global concern over the long-term availability of nonrenewable mineral fertiliser resources, and over the substantial energy consumption and greenhouse gas emissions from conventional N fertiliser production. Biosolids produced from treatment of domestic sewage has long been applied to agriculture as a source of nutrients to displace mineral fertilisers. For example, biosolids use in Australian agriculture has seen a 73 % increase in total production from 2010 to 2021. Significant Australian studies, such as the National Biosolids Research Program (2002–2008), have highlighted the potential FRV, crop returns and overall benefits and risks of biosolids use in agriculture. However, although the direct agricultural recycling of biosolids is still the most practicable option for beneficial reuse, it poses several notable logistical, practical, environmental and performance difficulties. These include (1) an imbalance of fertiliser nutrients in biosolids, and properties of biosolids that do not enable wellcontrolled field-application, with risk of over-supply or under-supply of nutrients and associated financial and environmental risks; and (2) emerging contaminants such as micro-plastics and nano-plastics which are ubiquitous in biosolids and could pose a significant future threat to the direct agricultural use of biosolids. Instead, thermal processing (e.g. incineration, pyrolysis or gasification) of biosolids has been explored to destroy such organic contaminants. Policy guidance regarding the safe use of biosolids in Australian agriculture has been well established and has been successful for >20 years to reduce potential adverse human health and environmental impacts associated with its beneficial use. For example, via a collation of data from several Australian literature studies, the current work demonstrates that heavy metal concentrations in biosolids have progressively declined over time, likely at least partly influenced by tighter control over industrial catchments. Due to recent scientific advancements on the role of soil microbiota on soil health, regulations for biosolids application to land could into the future consider the impact of contaminants (i.e., heavy metals, organic pollutants and microplastic) on the soil microbiota by the implementation of ecotoxicological analysis on soils.

As an alternative to destructive dissipation of potentially valuable carbon and nutrients by thermal processing, the current paper highlighted commercially-available extraction technologies as an alternative to instead recover such constituents from sewage sludge or biosolids upstream in the wastewater treatment plant, to make these available for agronomic use. However, although commercially available and applied in isolation, extractive technologies need to be researched and developed in an integrated whole-of-plant context and at a relevant scale, including to address key challenges such as a current high energy demand of N recovery, and a generally high comparative cost of extractive recovery.

Nutrients and carbon made available by extraction, put together with by-products from thermal processing (e.g. biochar), represent potential ingredients for the future production of balanced organic fertilisers, whether these be blends of mineral fertilisers and organic fertilisers, or whether instead reactive mixtures of these as formed, referred to as OMF. The understanding and further development of such products will be important to address current limitations, including variable nutrient mineralisation, release and supply, but also to provide fertiliser products suitable for broadacre cropping applications. For example, conventional spreading equipment used for mineral fertilisers with appropriate particle size and strength. Moreover, there are particular opportunities to balance the rapid plant-available nutrient supply of mineral fertilisers with slow-release organic fertiliser forms to provide sustained nutrient supply for enhanced crop growth. This represents a research opportunity and need to develop tailored fertiliser products with the desired product and nutrient supply characteristics. Research and investigations should particularly seek to resolve important links between processes that produce and extract nutrient and carbon constituents, and the agronomic benefits and risks posed when these constituents are formulated into targeted fertilisers and, importantly, applied to land at reasonable/viable application rates. This development could ensure the safe and beneficial use of nutrient and carbon resources in sewage sludge and biosolids across broad-acre agriculture into the future.

CRediT authorship contribution statement

Conceptualization B.K.M, S.M. S.T. and D.L.A.; methodology, S.M., S.T., and B.K.M.; formal analysis, S.M., S.T, P.S., and P.H.; investigation S.M and S.T.; data curation, S.M.; writing—original draft preparation, S.M.; writing —review and editing, S.M., S.T., P.S., P.H., D.L.A., and B.K.M.; visualization, S.M., S.T., D.L.A and B.K.M. supervision, B.K.M., project administration, B.K.M.; funding acquisition, B.K.M. All authors have read and agreed to the published version of the manuscript.

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Data availability

No data was used for the research described in the article.

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.

Appendix A. Supplementary data

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