The Impact of Banding Polymer-Coated Urea on Nitrogen Availability and Distribution in Contrasting Soils

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Abstract
Polymer-coated urea (PCU) has been traditionally used for broadcast and/or incorporated application of nitrogen (N) fertilizers. To improve N use efficiency (NUE), there has been an increase in sub-surface banded application of this fertilizer technology. However, there is little information on the release and supply of N from PCU granules when applied in a band. This research aimed to investigate the spatial distribution of key N transformations around PCU bands in soils of contrasting physico-chemical properties, and the implications for NUE. Two experiments, consisting of a 60-day diffusion study and a 91-day incubation, were conducted in a Vertosol and Dermosol, with PCU granules banded at a rate equivalent to 150 kg N ha⁻¹ and band spacing of 1.8 m. Compared to standard urea, PCU provided a sustained release of urea-N to soil solution and the lower urea-N concentrations minimized the toxic conditions associated with rapid hydrolysis of urea-N. Nitrogen release from banded PCU was quicker in the Vertosol (cf. Dermosol), possibly due to a higher volumetric water content and/or greater soil particle surface contact, facilitating rapid water imbibition into granules. However, the proximity of PCU granules to each other in a band restricted the diffusive release of urea-N from PCU granules (cf. dispersed application in both soils. Furthermore, the relatively mild chemical conditions in the PCU band (cf. standard urea) resulted in oxidation of larger proportions of PCU-derived mineral N. Banded application may extend the duration of N release from PCU granules, confounding efforts to predict N availability. Soil characteristics influenced N release and dynamics from banded PCU, although further investigation is required. Higher rates of nitrification of N derived from PCU bands suggest there could be increased risk of N-loss via denitrification or leaching pathways (cf. standard urea bands) if release dynamics are not optimally synchronised with crop demand. This study provides the first mechanistic insights into the impact of application method and soil physico-chemical properties on the efficacy of PCU.

Keywords Controlled release · Fertilizer · Coatings · Nitrogen use efficiency · Enhanced efficiency fertilizers · Soil fertility

1 Introduction
Nitrogenous fertilizers have sustained an estimated 40% of the global population since the late twentieth century by way of increases in agricultural productivity (Erisman et al. 2008; Pikaar et al. 2017). However, a significant side-effect of nitrogen (N) fertilizer application is the adverse impacts of off-site losses to surrounding ecosystems (Hatfield and Follett 2008). Enhanced efficiency fertilizers (EEFs) are proposed as a key tool for improving N use efficiency (NUE) in agriculture by improving the availability of N for crop uptake. Controlled release fertilizers (CRFs) represent an important class of EEFs whereby the rate, pattern and duration of N release are regulated under known conditions (Shaviv 2001). The object of controlling the release of N from granules is to better synchronize supply of N to
crop demands, minimising the opportunities for N loss in response to changing environmental conditions. Controlled release fertilizers are therefore designed for use via a single application that will progressively deliver crop N requirements over a growing season.

Typically, N-based CRFs are urea-N cores coated with materials such as sulphur and wax, polymer-sulphur, thermoplastics and resin polymers. However, these products have been variable in their effectiveness. Sulphur-coated urea has been largely replaced by newer coating technology because of an ineffective coating process that compromised the integrity of the coat and resulted in only ca. 30% of the product performing as expected (Shaviv 2001; Trenkel 2010). Polymer sulphur–coated products have a characteristic rapid initial release of over 20% of the total N content and a substantially delayed release of over 30% (Raban 1994), which makes them unsuitable for short season agricultural production, but popular in turf, landscaping and perennial horticulture markets. Thermoplastic-coated products consist of a nutrient coated in thermoplastic materials dissolved in a chlorinated hydrocarbon (Fujita 1995). The release of nutrients along concentration gradients from these products is temperature dependent, with moisture having a moderate effect, but the release rate is largely unaffected by pH and soil microbial activity (Christianson 1988). Resin polymer–coated fertilizers employ resin coatings which are hydrophobic and typically degrade with increasing temperature (Shaviv 2001). Two main types of resins are used as follows: alkyd-type resins and polyurethane-like coatings, which form a ‘skin-like’ layer on urea granules (Li et al. 2012). These resin-coated fertilizers release nutrients by allowing delayed water penetration through microscopic pores, thereby increasing osmotic pressure within the coated granule. As the coating is stretched, the micro-pore diameters increase, allowing nutrient release to soil solution along a concentration gradient (Azeem et al. 2014; Shaviv et al. 2003). The rate of release from these products is largely dictated by varying coating thicknesses and the composition of the resin (Shaviv 2001; Trinh et al. 2014).

The superior durability of thermoplastic and alkyd-type resin coatings in response to most environmental variables (i.e., moisture, microbial digestion, pH, etc.) means this technology is more predictable in its particular release patterns and has come to dominate the commercial market in recent years (Trenkel 2010). However, the manufacturing standard for determining N release patterns typically involves placing a single granule in a water solution at a known temperature and quantifying the change in N concentration in water over time (Azeem et al. 2014; Shaviv et al. 2003). This does not replicate the conditions that granules are likely to encounter when placed in, or on, soil in field situations. Consequently, matching the predicted release patterns with actual crop demand and nutrient acquisition can often produce unexpected outcomes. This is reflected in the mixed results reported from field trials investigating the effectiveness of polymer-coated urea (PCU) against a range of criteria such as crop yield, N2O emissions and ammonia volatilization (Martinez et al. 2021; Maharjan et al. 2017; Wang et al. 2016). While variability between sites and studies has been attributed to soil and climatic variation, it is possible that fertilizer placement (i.e., banding vs broadcasting) may also contribute to the variability in reported effectiveness of PCU. Janke et al. (2019) raised this concern when investigating the efficacy of PCU when applied in concentrated sub-surface bands, due to the reliance of these products on the development of strong concentration gradients between the solution inside the PCU granules and the surrounding soil solution. Incomplete release of urea from banded PCU in that study was attributed to neighbour-granule effects that reduced localised concentration gradients and subsequent diffusion rates, thereby slowing the release of urea to an extent that may limit plant N supply at times of peak demand. While the study by Janke et al. (2019) provided interesting insights, only the fertosphere (i.e., spatial volume comprising the fertilizer band and soil within 0.01 m of the fertilizer band) effects were investigated, and there was no consideration of the distribution of N beyond this zone.

There is therefore a need to characterize the performance of PCUs in soil and determine the efficacy of this technology in banded applications. The experiments described in this paper aim to (i) investigate the release and distribution of N within, and at distances from, the fertosphere of banded PCU (alkyd-type resin coating) in two contrasting soil types; and (ii) to identify if banding influences release characteristics of PCU granules compared to dispersed fertilizer applications. This was undertaken through two experiments. First, a diffusion study (designated ‘diffusion experiment’) examined the biochemical changes and N distribution in and around bands of PCU in soils with differing physico-chemical properties and where appropriate, compared changes to those from a standard urea band. Second, an extended incubation experiment provided a direct comparison of N release from dispersed vs banded PCU in a single soil type, to confirm the existence of a ‘neighbour-granule’ effect under banded conditions (designated as ‘fertilized soil volume experiment’).

2 Materials and Methods

2.1 Soil Collection, Characterization and Preparation

Soil from the top 10 cm of the profile of a yellow Dermosol and a black Vertosol (Isbell 2016) was collected from sugarcane farms in the Bundaberg district (Queensland,
of a N-fertilizer band applied at 150 kg N at a rate equivalent to the weight of fertilizer in a 5 cm length of the centre of the pot in an excavated cavity (1 cm diameter). Air-dried soil. Soil was air-dried and sieved to <2 mm. The moisture content at field capacity for each soil was determined using the column method described in Asher et al. (2002) and adjusted to take into account moisture held in air-dry soil.

### 2.2 Diffusion Experiment

#### 2.2.1 Experiment Design

Air-dried (sieved <2 mm) Dermosol and Vertosol (Table 1) were placed in 225 mm diameter PVC dishes to a depth of 5 cm and packed to bulk densities of 1.56 and 1.45 g cm⁻³, respectively (ca. 3100 g Dermosol, 2888 g Vertosol per pot). Deionized water was added to each pot to bring the soil mass to the pre-calculated field capacity moisture content (0.12 g g⁻¹ and 0.32 g g⁻¹ for the Dermosol and Vertosol, respectively). All pots were sealed with plastic cling wrap and allowed to equilibrate overnight. It is noted that following moisture application, the Vertosol swelled approximately 0.5 cm in height, reducing the bulk density of this soil to ca. 1.32 g cm⁻³. Fertilizer N treatments were then applied into the centre of the pot in an excavated cavity (1 cm diameter) at a rate equivalent to the weight of fertilizer in a 5 cm length of a N-fertilizer band applied at 150 kg N ha⁻¹ at 1.8 m row spacing (270 mg N cm⁻¹ band length). Once applied, the exposed end of the N-fertilizer band/column was covered with some of the previously removed soil to limit the chance of volatilization losses. The N-fertilizer treatments applied in this experiment included the following: (i) standard granular urea; and (ii) PCU granules (Agromaster Tropical®) with a reported release period of 90 days, determined by the industry standard method of dissolution in water at a constant temperature. Untreated soil of both the Vertosol and Dermosol was also included as a negative control and results for this treatment are presented in Part A of the Supplementary Material. A moist filter paper (Whatman® 5C, 125 mm diameter) was placed centrally on top of the soil to maintain humidity within the pot and allow for moisture adjustment without disturbing soil. Incubation pots were weighed to allow for on-going moisture adjustment, with moisture maintained at field capacity ±5% (moisture adjustment every 7 days). Incubation pots were sealed with plastic cling wrap and three pinholes were inserted in the cling wrap of each incubation pot to prevent the development of anaerobic conditions. The incubation was maintained at 25 ± 2 °C over 60 days and triplicate pots were placed in a completely randomised design across two benches.

#### 2.2.2 Sample Collection

At 2, 10, 20, 35 and 60 days after incubation initiation (DAI), soil in each pot was collected from a 2 cm diameter central core (designated the ‘0–1 cm’ position or ‘fertosphere’). Sampling continued in 2 cm increments moving outwards from that central core using stainless steel sampling rings of diameters 4, 6, 8 and 10 cm, representing zones 1–3 cm, 3–5 cm, 5–7 cm, 7–9 cm and 9–11 cm from the fertosphere, respectively. Soil in each increment was completely removed from incubation pots and mixed to homogenize.

#### 2.2.3 pH and Electrical Conductivity Measurements

The pH and electrical conductivity (EC) of soil samples were measured in a 1:5 (w/v) soil–water solution by adding 50 mL DI water to 10 g soil from each distance zone. The pH and EC were measured using a pH/EC Robot (Labfit AS-3020) with 45 s stirring prior to instrument reading. The pH was recorded when multiple (5) values within a range of 0.02 were recorded, and likewise EC values were taken when variation between 5 readings was within 0.02 dS m⁻¹. A maximum reading time length of 60 s was set for the pH and EC of each sample.

### Table 1 Key soil chemical properties and characteristics of the surface 0–10 cm. EC, electrical conductivity; pHBC, pH buffering capacity; CEC, cation exchange capacity; TC, total carbon; TOC, total organic carbon; TN, total nitrogen; NH₄⁺-N, ammonium nitrogen; NO₃⁻-N, nitrate nitrogen

| Soil order | pH 1:5w | EC 1:5 | pHBC | Particle size analysis (%) | CEC | TC | TOC | TN | NH₄⁺-N | NO₃⁻-N |
|------------|---------|--------|------|---------------------------|-----|----|-----|----|--------|--------|
|            | dS m⁻¹  | cmol OH-kg⁻¹ | pH unit⁻¹ | Coarse sand | Fine sand | Silt | Clay | cmol(+) kg⁻¹ | % | mg kg⁻¹ |
| Dermosol   | 6.30    | 0.07   | 1.32 | 38 | 46 | 9 | 9 | 4.5 | 1.21 | 1.17 | 0.06 | 3.2 | 6.8 |
| Vertosol   | 7.15    | 0.35   | 3.51 | 5 | 26 | 21 | 52 | 28.5 | 2.36 | 2.32 | 0.18 | 4.8 | 8.5 |

¹Isbell 2016
2.2.4 Soil Extraction

Extraction of mineral N followed standard methods (method: 7C2a; Rayment and Lyons 2010), using a 1:5 (w/v) soil-solution extraction with 2 M potassium chloride (KCl) solution (i.e., 10 g soil with 50 mL 2 M KCl). A 10 mL aliquot of the extraction solution was taken and centrifuged at 3700 rpm (RCF: 1760 × g) for 9 min, followed by pouring off and re-centrifuging of the supernatant. All solutions were frozen at –80 °C until analysis. The solutions were ing off and re-centrifuging of the supernatant. All solutions were expressed as the solution concentration of extrac-
tions (i.e., soil solution concentration). Mineral N val-
ues are reported as mg N kg⁻¹ soil−1 as these data were obtained from soil extracts (1:5 w/v 2 M KCl) and the mg N kg soil−1 unit is comparable to that used in other studies. Concentrations of NO₃⁻⁻N were determined by calculating the difference between NO₃⁻⁻N and NO₂⁻⁻N concentrations (Best, 1976; Bremner, 1965). Ammonia (aqueous NH₃) concentrations (mg L⁻¹) were calculated by applying the pKₐ value of the NH₄⁺ ↔ NH₃ reaction at 25 °C (Emerson et al. 1975) to a rearranged version of Eq. 1 in Erickson (1985) and converting the moles-in-
solution output by multiplying by the molecular weight of NH₃. Values for pH and EC were arithmetically averaged from triplicate samples. Heat maps were used to display changes over time (treatment value minus control value) for pH and EC, with respect to distance from the fertilizer band. The figure key indicates the colour and intensity of each zone which correlates to a level of change for pH and EC. Statistical analysis of chemical data (pH and EC) and N (urea-N, mineral N and aqueous NH₃) concentrations was conducted by a three-way analysis of variance (ANOVA) for each soil with time, treatment (N-fertilizers) and distance from fersphere as factors. Tukey’s honestly significant difference (HSD) test (P < 0.05) was used to test means for significant interaction effects. For urea-N, mineral N and aqueous NH₃ datasets, statistical variation is represented in figures by standard error bars fitted to mean values at each point. Statistical differences for pH and EC are not presented within the manuscript as the number of interactions makes graphical presentation difficult to interpret. Where appropriate, significant effects are noted in the text, with the full statistical results available in Supplementary Material (Part C).

2.2.5 Data Transformation and Statistical Analyses

Urea-N (mg urea-N L⁻¹) values from U-HPLC analysis were expressed as the solution concentration of extrac-
tions (i.e., soil solution concentration). Mineral N values are reported as mg N kg soil−1 as these data were obtained from soil extracts (1:5 w/v 2 M KCl) and the mg N kg soil−1 unit is comparable to that used in other studies. Concentrations of NO₃⁻⁻N were determined by calculating the difference between NO₃⁻⁻N and NO₂⁻⁻N concentrations (Best, 1976; Bremner, 1965). Ammonia (aqueous NH₃) concentrations (mg L⁻¹) were calculated by applying the pKₐ value of the NH₄⁺ ↔ NH₃ reaction at 25 °C (Emerson et al. 1975) to a rearranged version of Eq. 1 in Erickson (1985) and converting the moles-in-
solution output by multiplying by the molecular weight of NH₃. Values for pH and EC were arithmetically averaged from triplicate samples. Heat maps were used to display changes over time (treatment value minus control value) for pH and EC, with respect to distance from the fertilizer band. The figure key indicates the colour and intensity of each zone which correlates to a level of change for pH and EC. Statistical analysis of chemical data (pH and EC)

2.3 Fertilized Soil Volume Experiment

2.3.1 Experiment Design

Exactly 500 g of air-dry Dermosol (sieved to <2 mm) was placed in round plastic pots (ca. 90 mm diameter ×90 mm height) and 60 mL of DI water was added to bring the soil to field capacity (0.12 g g⁻¹). Screw-lids containing a cen-
tral pinhole for gas exchange were fastened onto the plastic pots and the soil plus water were allowed to equilibrate overnight. Fertilizer treatments were applied by either (a) removing a central core of soil and filling the cavity with PCU granules (banded treatment), or (b) placing soil into a large tray and mixing with granules before placing back in the pot (dispersed treatment). Treatments included the fol-
lowing: (i) control (untreated), (ii) standard urea banded, (iii) standard urea dispersed, (iv) PCU banded and (v) PCU dispersed. Fertilizer treatments were applied at an in-
band concentration equivalent to a rate of 150 kg N ha⁻¹ at 1.8 m spacing (i.e., 1350 mg urea-N pot⁻¹). Individual pots were then weighed as a benchmark so that soils could be maintained at field capacity ±1% (moisture adjustment every 7 days) for the duration of the study. The incubation was maintained at 25 ± 2 °C and triplicate pots were placed in a completely randomised design on a laboratory bench.

2.3.2 Sample Collection

At the time of sampling (7, 14, 28, 56 and 91 DAI), the contents of each pot were placed in a tray and the indi-
vidual PCU granules were carefully recovered from the soil. The soil was subsequently mixed and a 100 g sample placed into a 1 L bottle for extraction with 2 M KCl. A separate 10 g soil sample was placed in a 70 mL specimen container for pH and EC analyses.
2.3.3 Soil extraction, pH and Electrical Conductivity

Measurement of pH and EC followed procedures outlined for the diffusion experiment (Sect. 2.2.3) using 10 g of soil. Extraction and measurement of mineral N from soil samples followed protocols outlined in Sect. 2.2.4 using 100 g of soil.

2.3.4 Recovery and Analysis of Urea in PCU Granules

The granules recovered from soil were crushed using a mortar and pestle and re-suspended in 20 mL of DI water to which a urease inhibitor (10 µL of 0.02 M Ag$_2$SO$_4$) was added. Approximately 5 mL of solution was filtered to 0.45 µM to remove polymer residues and soil particles and then frozen until analysis by HPLC. Urea-N was determined following procedures outlined in Sect. 2.2.4. Data were processed using the Chromeleon 7.4 acquisition software and results were reported as mg L$^{-1}$.

2.3.5 Data Transformation and Statistical Analysis

In this fertilized volume experiment, urea-N recovery was calculated as the proportion (%) of total applied urea-N detected in water extracts (20 mL) from recoverable PCU granules. Mineral N (NH$_4$-N + NO$_x$-N) is reported as absolute values (mg N), with concentrations of NO$_x$-N determined by calculating the difference between NO$_3$-N and NO$_2$-N (Best 1976; Bremner 1965). Statistical analysis of urea-N and total mineral N was conducted by two-way ANOVA comparing the mean differences of time and treatment (N-fertilizer × application method), followed by post hoc Tukey HSD analysis ($P < 0.05$).

3 Results

3.1 Diffusion Experiment

3.1.1 Urea-N

Urea-N was largely confined to the fertosphere (0–1 cm from the fertilizer band) of PCU bands in both the Dermosol and Vertosol over 60 DAI, with concentrations fluctuating over time (Fig. 1). Except at 10 DAI, PCU in the Vertosol generally exhibited higher urea-N concentrations in the fertosphere cf. the Dermosol, although concentrations between soils were not directly compared using statistical methods. In contrast, urea-N from urea bands was only detected at 2 DAI but elevated concentrations extended as far as the 1–3 cm zone in both soils (Fig. 1).

3.1.2 NH$_4$-N and NO$_3$-N

The composition of mineral N in both soils treated with banded PCU was dominated by NH$_4$-N (Fig. 2a, b). While this was also the case for urea bands, soil solution around PCU bands never became as concentrated and peak NH$_4$-N concentrations (ca. 765 mg N kg$^{-1}$ soil for Dermosol and...
2470 mg N kg⁻¹ soil for Vertosol) occurred much later (ca. 33 days) compared to standard urea. Dispersion of PCU-derived NH₄-N was similar in both soils over time, although concentrations were 48% higher in the 0–3 cm zone of the Vertosol by 60 DAI. Significant increases in NO₃-N formation were observed from 20 DAI within the fertosphere and up to 5 cm away in both soils (Fig. 2a, b). While initial formation of NO₃-N from PCU bands (i.e., at 20 DAI) was similar in both soils, concentrations appeared to increase somewhat more rapidly over time in the Vertosol (19–28% higher in the 0–5 cm zone). These dynamics contrasted to that of standard urea where initial NH₄-N concentrations were significantly elevated and decreased slowly over time and NO₃-N increased significantly in zones further from the fertosphere, viz. the 1–7 cm and 5–9 cm zones in the Vertosol and Dermosol, respectively (Fig. 2a, b).

3.1.3 Aqueous NH₃

Banding PCU produced much lower quantities of aqueous NH₃ cf. standard urea in both soils, although concentrations were higher (peak of 14 mg L⁻¹ in Vertosol vs 2 mg L⁻¹ in Dermosol) and more variable in the Vertosol (Fig. 3). In contrast, bands of standard urea immediately (i.e., evident at 2 DAI) produced high aqueous NH₃ concentrations within the fertosphere of the Vertosol (32 mg L⁻¹) and up to 3 cm from the fertosphere in the Dermosol (13 mg L⁻¹; Fig. 3). Aqueous NH₃ dissipated over time, although the decline was more rapid (ca. 15 days) in the Dermosol than the Vertosol.

3.1.4 pH of Soil Solution

Where rapid increases of ca. 2.8 pH units were observed in and around urea bands from 2 DAI, a significant increase
was not observed for PCU bands until 10 DAI (1.86 pH units, Fig. 4). The zone of the greatest pH change was largely restricted to the fertosphere of PCU bands and pH remained elevated (ca. 8.45) in this zone for the entire 60 days in both soils. In contrast, significant pH increases were observed up to 4 cm and 7 cm from urea bands in the Vertosol and Dermosol, respectively, although pH increases at distance from the fertosphere had begun dissipating by 20 DAI in both soils (Fig. 4). This acidification was first observed in the outer 7–11 cm of both soils treated with banded urea, with the magnitude of pH decline increasing and the affected zone expanding to include soil solution closer to the fertosphere over time (Fig. 4). The extent of acidification and the volume of soil acidified was greater around urea bands in the Vertosol, with decreases of 0.78–1.62 pH units in the 3–11 cm zone compared to the Dermosol (decline of 0.99–1.29 pH units in the 5–11 cm zone) by 60 DAI. However, in soils treated with PCU bands, acidification began earlier (ca. 15 days) and increased more rapidly (decline of 1.19–1.83 pH units by 60 DAI) in the Dermosol compared to the Vertosol (decline of 0.08–1.27 pH units). Notably, acidification of soil solution initiated much closer to the fertosphere of PCU bands (1–3 cm in the Vertosol and 3–5 cm in the Dermosol) compared to early pH declines in the outer zones around urea bands (Fig. 4).

### 3.1.5 Electrical Conductivity of Soil Solution

Significant increases in EC (ca. 0.59 dS m⁻¹) were not observed in soil treated with PCU until 20 DAI, with increases primarily localised to the fertosphere (Vertosol) and up to 3 cm away (Dermosol). In contrast, significant increases were seen from 2 DAI in the fertosphere (2.07 dS m⁻¹ in the Vertosol) and up to 3 cm (ca. 1.42 dS m⁻¹ in the Dermosol) from bands of urea. While increased EC did not disperse as far from PCU as for urea bands in both soils, there were differences in EC distribution between soils treated with PCU; dissolved soluble salts did not distribute as far from the fertosphere of PCU in the Vertosol (primarily with 3–5 cm) as for the Dermosol (5–7 cm). Results for EC distribution are presented in Part B of the Supplementary Material.

### 3.2 Fertilized Soil Volume Experiment

#### 3.2.1 Urea-N Retained in Granules

The retention of urea-N in PCU granules decreased significantly over time, regardless of whether granules were banded or mixed into soil (Fig. 5). However, while the release dynamics were similar regardless of application method, the total urea-N retained in granules was significantly greater when PCU was banded compared to dispersed through the soil volume at all stages of the 91-day incubation. Differences were initially small at 7 DAI (2% difference in urea-N release), but steadily increased thereafter such that the amount of urea-N retained in granules by 28 DAI was almost 10% lower in the dispersed than the banded treatment. The difference remained between 5

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Fig. 3 Formation and distribution of aqueous ammonia (NH₃; mg L⁻¹) over 60 days after incubation (DAI) in response to the addition of bands of either standard urea or polymer-coated urea in the Dermosol (a–e) and Vertosol (f–j). Aqueous NH₃ in untreated soils was always <0.0001 mg L⁻¹. The fertosphere comprises a spatial volume of soil up to 1 cm from the band location and is represented at ‘0’ in the figure. Subsequent zones were sampled in 2 cm increments, concentrically. Vertical bars represent the standard error of the means presented for each time × location. Full statistical output for each soil with time, treatment and distance from the fertosphere as factors of a three-way ANOVA, followed by post hoc Tukey’s HSD analysis, is located in Supplementary Material Part C.
and 10% until the experiment was terminated at 91 DAI—the reported release timeframe for this PCU product at 25 °C. At this time, only 83% and 78% of the urea-N had been released from dispersed or banded PCU granules, respectively.

3.2.2 \( \text{NH}_4^+\text{-N and NO}_3^-\text{-N} \)

The quantity of mineral N released from PCU granules and measured in the soil increased over time in both PCU treatments. This pattern contrasted sharply with that of urea, which decreased rapidly from an initial peak at the first
sampling occasion (7 DAI, Fig. 6). In both PCU treatments, both NH$_4$-N and NO$_3$-N increased slowly and maintained fairly equal proportions throughout the incubation period (Fig. 6). In contrast, the mass of mineral N in the urea treatment declined to ca. 60% of the maximum by 91 DAI and was almost exclusively present as NH$_4$-N (average of 99%) during the entire incubation.

In the PCU treatments, significantly more mineral N was recovered over the duration of the incubation in the dispersed treatment compared to that of the banded. This was most notable from 14 DAI, when 160 mg of mineral N was measured in the dispersed treatment compared to 113 mg in the banded (30% lower), with the magnitude of this difference increasing to 53% (454 cf. 215 mg) at 28 DAI. However, while a trend for lower mineral N in the banded treatment was maintained until the end of the incubation, differences had reduced to less than 5% by 56 (744 cf. 714 mg, dispersed cf. banded) and 91 DAI (942 cf. 897 mg). Interestingly, measured mineral N at 91 DAI was still considerably less than the ca. 1000 mg (987 mg dispersed, 1014 mg banded) recorded in the urea treatments at 7 DAI (Fig. 6). However, recovery of applied N from soil (mineral N) and PCU granules (urea-N) indicates that a significant proportion of the applied N remained within PCU granules throughout the incubation, although this decreased over time to ca. 20% (Table 2) at the end of the study. Notably, at all times, ca. 80% of applied N could always be accounted for in the PCU treatments whereas N recovery decreased from ca. 75% at 7 DAI to 45% at 56 DAI in the urea treatments (Table 2).

### 4 Discussion

Understanding CRF release patterns and subsequent N dynamics in soils is vital to optimising crop N uptake, thereby improving the efficacy of these products. The following discussion therefore focuses on release dynamics, and the major biochemical reactions in soil including (i) urea hydrolysis, (ii) the equilibria of NH$_4^+$ and NH$_3$ and (iii) nitrification, with all reactions referenced against a standard urea standard. The spatiotemporal dynamics of banded N, implications for crop NUE and mitigation of environmental N losses are also incorporated into this discussion. The effects of banding (cf. dispersed applications) are assessed from the fertilized soil volume experiment while the impact of key soil physico-chemical properties is assessed from results of the diffusion study.

This study finds that banded fertilizer application will influence the release profile of N from PCU where the release of urea-N from banded PCU granules was significantly lower than that of equivalent dispersed applications over 91 DAI (Fig. 5). This is consistent with the hypothesis of Janke et al. (2019) that suggested smaller concentration gradients between granules and the surrounding soil solution develop when granules are applied in a band, compared to dispersed through the soil volume. This hypothesis is formed on the premise of diffusion-based urea-N release from polymer-coated granules following initial water penetration through microscopic pores in the polymer-coating. As osmotic pressure within the granule increases, these micropores are stretched and N is released along a
concentration gradient to the soil solution (Azeem et al. 2014; Shaviv et al. 2003). In a fertilizer band, the centre-to-centre distance between PCU granules is of the order of ca. 4 mm, given the average PCU granule diameter of 3.88 mm. In contrast, the centre-to-centre distance between dispersed PCU granules in this incubation was calculated as ca. 18.7 mm, on the basis of the average PCU granule volume, the volume of soil and the number of granules applied. The local N concentration surrounding individual PCU granules in a band is therefore likely to be much higher, consequently lowering the granule-soil solution concentration gradient and diffusive potential for N release (cf. mixed applications).

When granule density is further reduced in even larger soil volumes (as would occur under field conditions), the mean inter-particle distance of dispersed PCU granules will be much greater than in this experiment, whereas the proximal distance of banded granules would likely be similar to that in this experiment. This will result in even greater contrasts between the PCU-N release dynamics of banded vs dispersed PCU granules.

While N retained in PCU granules was not measured in the diffusion study, the dynamics of urea-N in the soil solution (PCU cf. standard urea) confirmed the delayed release of urea-N from these products (Fig. 1). Furthermore, differences in urea-N concentrations observed in close proximity to N-fertilizer bands (Fig. 1) were indicative of the effects of key physico-chemical soil parameters on both the rate of urea-N release from granules and the rate of urea hydrolysis. Greater differences in urea-N concentrations around bands of PCU than those of standard urea in the Vertosol (cf. the Dermosol; Fig. 1) are consistent with the higher clay and organic carbon (OC) content in the Vertosol potentially supporting greater ureolytic activity and more rapid hydrolysis of urea-N. Urease is the enzyme which facilitates hydrolysis of urea-N, and activity of this enzyme in soils is positively correlated with both clay and OC content (Zantua and Bremner 1977). This hypothesis is supported by greater formation of NH₄-N around standard urea bands in the Vertosol cf. Dermosol, indicating greater ureolytic activity in this soil (Fig. 2). When PCU bands were
Slower release of N from PCU meant that NH$_4$-N concentrations did not peak as high as those surrounding standard urea, despite adequate rainfall/irrigation (Janke et al. 2020; Martinez et al. 2021). These findings also highlight the risk of N deficiencies from CRFs under dry soil conditions. Nitrogen ‘stranding’ in topsoil layers (i.e., the part of the profile where N-fertilizer is applied) that have dried out during the growing season is an established issue in agricultural systems, resulting in crop roots being unable to access the applied N (e.g., dryland systems; Armstrong et al. 2015). This issue may be exacerbated for CRF technologies where N release may be impeded by both limited penetration of water into granules and by restricted diffusive pathways out into soil solution. In terms of improving crop NUE, the synchrony of N release from CRFs with plant N demand is also critical, with long-season crops which have an extended uptake period demonstrating better responses to the delayed release of N (e.g., Rathnappriya et al. 2022 cf. Martinez et al. 2021, Schwenke and Haigh 2019). Since banded applications may exacerbate the delayed release of N from CRF products, the mode of fertilizer application may also have an impact on how effectively N supply is synchronised to plant demand. Thus, while the exact process(es) driving differences in N release from PCU remain unclear, it is evident that soil moisture and texture variables which influence the formation of N concentration gradients around PCU granules are likely key factors driving release dynamics. These factors are therefore expected to influence crop NUE although this aspect was not specifically investigated in this study.

Slower release of N from PCU meant that NH$_4$-N concentrations did not peak as high as those surrounding standard

| Table 2 | Recovery (%) as total nitrogen (N) of applied N (bold values) ± the standard deviation in soil and polymer-coated urea (PCU) granules in the Dermosol from 7 to 91 days after incubation (DAI) in the fertilized soil volume experiment. Fertilizer treatments were applied by either mixing into soil (mixed) or in central bands (banded) within incubation pots |
|----------|-----------------|-----------------|-----------------|-----------------|
|          | Urea mixed      | Urea banded     | PCU mixed       | PCU banded      |
| 7 DAI    | Soil 74 ± 1     | 76 ± 2          | 4 ± 0           | 3 ± 0           |
|          | Granule         | -               | 73 ± 2          | 75 ± 3          |
|          | Total 74 ± 1    | 76 ± 2          | 77 ± 0          | 78 ± 3          |
| 14 DAI   | Soil 67 ± 1     | 71 ± 1          | 12 ± 2          | 8 ± 1           |
|          | Granule         | -               | 71 ± 0          | 76 ± 1          |
|          | Total 67 ± 1    | 71 ± 1          | 83              | 84 ± 2          |
| 28 DAI   | Soil 59 ± 1     | 61 ± 4          | 34 ± 0          | 26 ± 2          |
|          | Granule         | -               | 50 ± 0          | 57 ± 1          |
|          | Total 59 ± 1    | 61 ± 4          | 84              | 83 ± 3          |
| 56 DAI   | Soil 44 ± 1     | 47 ± 3          | 55 ± 3          | 53 ± 2          |
|          | Granule         | -               | 26 ± 1          | 32 ± 1          |
|          | Total 44 ± 1    | 47 ± 3          | 81              | 85 ± 3          |
| 91 DAI   | Soil            | -               | 70 ± 1          | 66 ± 1          |
|          | Granule         | -               | 17 ± 1          | 22 ± 1          |
|          | Total 87 ± 2    | 88 ± 2          |

applied, it is suggested that the higher clay and volumetric water content at field capacity in the Vertosol enabled better contact between soil particles and their surrounding water shells with PCU granules. This facilitated more rapid imbibition of water into PCU granules, enabling the quicker release of urea-N into soil solution by 2 DAI (cf. Dermosol; Fig. 1). In contrast, the sandier texture and lower volumetric water content of the Dermosol is unlikely to have been as effective at facilitating water imbibition and subsequent urea-N release from PCU granules, resulting in the significantly lower urea-N concentrations observed throughout the incubation (Fig. 1). This hypothesis is consistent with recent findings (Verburg et al. 2021) that soil moisture and texture variables play an important role in facilitating N release from PCU granules due to their influence on water uptake. Earlier studies by Cahill et al. (2010) and Golden et al. (2011) also indicated that higher soil moisture contents are likely to facilitate earlier and greater release of N from PCU products. Although urea-N retained in PCU granules and enzymatic activity were not directly measured in this experiment, the impact of initial water imbibement on subsequent release appears to be the dominant process controlling urea-N concentrations in soil solution surrounding banded PCU. Furthermore, although Golden et al. (2011) note that soil water content had little impact on N release once the release process had been initiated, conditions which prevent localised N accumulation and facilitate movement of N away from the band zone are likely to enhance diffusion-based N release by way of increasing concentration gradients. For example, in scenarios where there is substantial mass flow through PCU bands (e.g., rainfall and irrigation events in soil profiles that facilitate hydraulic movement), N may be more widely distributed throughout the soil profile, enabling formation of greater concentrations gradients and, consequently, more rapid and complete N release. Enhanced N release from PCU granules in response to dynamic water movement (cf. static conditions) has been previously demonstrated in water-based studies and attributed to formation of greater N concentration gradients between intra and outer granule solutions as a result of removal of released N (Shen et al. 2015). The availability of sufficient soil moisture and soil properties which are conducive to mass flow is therefore likely to be important for effective N release from banded CRF products. It is speculated that these factors may also improve crop N acquisition from banded PCU by facilitating a wider zone of N distribution and thereby providing a larger volume of N-enriched soil in which plant roots may access fertilizer N. Soil characteristics may be particularly influential, with recent studies of N movement from fertilizer bands in a high clay and CEC soil demonstrating considerably smaller N-enriched soil volumes around CRF bands (cf. standard urea), despite adequate rainfall/irrigation (Janke et al. 2020; Martinez et al. 2021). These findings also highlight the risk of N deficiencies from CRFs under dry soil conditions. Nitrogen ‘stranding’ in topsoil layers (i.e., the part of the profile where N-fertilizer is applied) that have dried out during the growing season is an established issue in agricultural systems, resulting in crop roots being unable to access the applied N (e.g., dryland systems; Armstrong et al. 2015). This issue may be exacerbated for CRF technologies where N release may be impeded by both limited penetration of water into granules and by restricted diffusive pathways out into soil solution. In terms of improving crop NUE, the synchrony of N release from CRFs with plant N demand is also critical, with long-season crops which have an extended uptake period demonstrating better responses to the delayed release of N (e.g., Rathnappriya et al. 2022 cf. Martinez et al. 2021, Schwenke and Haigh 2019). Since banded applications may exacerbate the delayed release of N from CRF products, the mode of fertilizer application may also have an impact on how effectively N supply is synchronised to plant demand. Thus, while the exact process(es) driving differences in N release from PCU remain unclear, it is evident that soil moisture and texture variables which influence the formation of N concentration gradients around PCU granules are likely key factors driving release dynamics. These factors are therefore expected to influence crop NUE although this aspect was not specifically investigated in this study.
urea bands (Fig. 2a, b) and, consequently, the formation of concentration gradients between the PCU fertosphere and the soil further away was much lower (cf. urea treatments; Fig. 2). Interestingly, while the NH₄-N concentration gradient from PCU in the Vertosol was much greater than that in the Dermosol, this did not result in significantly differing zones of fertilizer NH₄-N enrichment (which occurred predominantly within 5 cm of the fertosphere in both soils; Fig. 2). This suggests that the high CEC in the Vertosol and the low tortuosity of the Dermosol collectively resulted in similar distributions of NH₄-N, despite absolute NH₄-N concentrations varying considerably (Fig. 2a, b). Since positively charged NH₄-N is relatively immobile in soil, it would be expected that factors such as soil texture (and dynamic water movement) would be less likely to impact NH₄-N distribution. However, NH₄⁺ exists in equilibrium with aqueous NH₃, with this reaction favouring formation of NH₃ as the pH of the surrounding solution increases (Robertson and Groffman 2015). It is therefore likely that at least part of the NH₄⁺-N around urea-based N-fertilizer bands dispersed as aqueous NH₃, thereby explaining how soil properties related to diffusion may impact NH₄-N distribution. It is also possible that aqueous NH₃ concentrations become so high that NH₃ volatilizes to a gaseous form across the soil-surface-air boundary layer (i.e., Eq. 2.3 in Creamer and Fox 1980), and moves through soil as a gas before redissolving into solution and converting to NH₄⁺ under lower pH and aqueous NH₃ conditions. A field trial by Rochette et al. (2013b) demonstrated this process, whereby NH₃ volatilization from concentrated sub-surface bands of standard urea increased NH₄⁺-N concentrations in the soil above fertilizer bands relative to that of untreated soil. A recent study (Hestrin et al. 2021) has also provided evidence for the importance of gaseous NH₃ in the transport and availability of N in soils. Since formation of aqueous NH₃ occurs in response to high pH and NH₄⁺-N concentrations (Emerson et al. 1975; Erickson 1985), it was not surprising that the greatest aqueous NH₃ concentrations were observed within and around urea bands (Fig. 3). Increases in pH are driven by the consumption of protons from soil solution during the hydrolysis of urea (Kissel et al. 1988), and NH₄⁺ is also formed during this ureolytic process. While these reactions are well established in the literature for standard urea, the impact of CRF technology on this chemical process is demonstrated in this study. The slower release of urea-N from PCU had the dual effects of minimising pH increases (Fig. 4) and maintaining lower NH₄⁺-N concentration (Fig. 2a, b), resulting in delayed (by 20–35 days) and lower peak NH₃ concentrations. Because changes in pH around PCU bands were relatively similar between soil types (Fig. 4), the dominant factor driving changes in aqueous NH₃ between the soils was the local concentration of NH₄⁺-N within the fertosphere. This indicates that use of PCU has the potential to significantly reduce the aqueous NH₃ toxicity and volatilization losses associated with banded urea (DeBruin et al. 2021; Ingle et al. 2010; Qin et al. 2014). Significant reductions in NH₃ volatilization have been previously reported for coated urea products (Lam et al. 2019; Lemonte et al. 2016; Rahman et al. 2018), although these studies primarily focused on surface broadcasting applications. Since sub-surface application of banded N-fertilizer may already mitigate a large portion of volatilization losses (Rochette et al. 2013a, 2013b), the benefit from PCU for conferring NUE improvements through reduced NH₃ emissions is likely to be minimal. However, several studies (Ingle et al. 2010; Qin et al. 2014) have demonstrated that use of PCU will minimise the potential for adverse impacts on crop establishment by reducing the concentrations of aqueous NH₃. Importantly, this study indicates that reductions in aqueous NH₃ and the associated phytotoxic effects are likely to be observed in many soil types, although the strength of the effect when compared to standard urea may vary (Fig. 3).

Nitrification within and around the banded N-fertilizers varied with treatment and soil type (Fig. 2). For standard urea, the delay in formation of NO₃-N until 10 DAI (Dermosol) or 20 DAI (Vertosol) was a consequence of raised pH, aqueous NH₃ concentrations and possibly increased EC collectively inhibiting nitrifying activity (Janke et al. 2021). As these conditions dissipated in the outer zones (Figs. 3, 4), nitrification commenced in those zones before gradually increasing in zones progressively closer to the fertilizer band over time (Fig. 2). However, much milder changes to soil chemistry (Figs. 3, 4), combined with limited movement of NH₄-N from the fertosphere, meant that nitrification was initiated within the fertosphere of PCU bands and increased in zones further from the band over time (Fig. 2). Thus, where differences in the zone of modified soil chemistry between soils influenced the zones of nitrification activity, soil properties did not significantly impact NO₃-N distribution around PCU bands (Fig. 2). While the predominant mineral N species was NH₄-N in all N-fertilizer treatments, the proportions of mineral N present as NO₃-N within PCU-treated soil were much higher in the 0–5 cm zone compared to urea treatments throughout both experiments (Figs. 2, 6). This is indicative of much higher rates of nitrification in PCU-treated soil and was particularly evident in the fertilized soil volume experiment (Fig. 6). Notably, concentrations of NO₃-N were higher for standard urea cf. PCU in the outer zone of the diffusion study (Fig. 2; 5–11 cm) in both soils, likely due to more extensive diffusion of aqueous NH₃ (Fig. 3), resulting in greater distribution of NH₄-N from the fertilizer band. It is therefore clear that soil chemical conditions (i.e., pH, EC and aqueous NH₃) do not become strongly inhibitory to nitrifying organisms (Anthonisen et al. 1976; Bae et al. 2001; Grunditz and Dalhammar 2001) within and around PCU bands. In this case, other soil factors such as
the soil matrix, redox status, temperature and intrinsic pH may become more important for regulation of nitrification (Sahrawat 2008). Under field conditions and compared to a standard urea band, rapid nitrification of released N from PCU suggests that a greater proportion of the mineral N present at any time may be susceptible to leaching or denitrification losses after heavy rainfall or large irrigation events. Indeed, several studies that directly compare sub-surface applications of PCU to standard urea have not found positive or conclusive evidence supporting increased NUE benefits from PCU by way of reduced NO$_3$-N leaching (Debruin et al. 2021; Janke et al. 2020; Martinez et al. 2021; Xie et al. 2020) or significant reductions in N$_2$O emissions (Dang et al. 2021; Rose et al. 2018). Of further interest is the observation of formation of significant NO$_3$-N concentrations at later stages of the incubation in the Vertosol treated with standard urea cf. the PCU (Fig. 2). This effect was accompanied by greater increases in acidity (Fig. 4), suggesting the initially inhibited nitrifier community recovered more quickly in the Vertosol. Nitrate formation and dissipation of alkaline conditions occurred to a lesser extent in the Dermosol over the 60-day incubation (Figs. 2, 4). This indicates that mitigation of N loss through the use of PCU may also be influenced by soil type, although further investigation is required. Of most importance, however, is the concern that the extended duration of release and subsequent nitrification of PCU-N may result in N loss pathways that can be activated over an extended period, creating a potentially large window for N loss risk. The concept of ‘whole of lifetime losses’ from fertilizer products has not been thoroughly investigated in the scientific literature and is a critical factor in determining whether CRFs can deliver NUE advantages and effectively mitigate environmental N losses.

This study was conducted in the absence of plants, which are an important sink for fertilizer N and a major driver influencing mineral N concentrations in different parts of the soil profile. Even in situations with high risks of N loss, the presence of an actively growing crop may reduce the risk of gaseous or leaching losses of NO$_3$-N by rapidly accumulating N in the crop biomass. In addition, there are clear effects of soil-fertilizer granule contact volume and sampling time reported in this paper that need to be considered in extrapolating results to different situations. The diffusion study was conducted over a shorter duration (60 days), in a larger volume of soil (ca. 3 kg) and was sampled in discrete zones at different distances from the fertilizer band. Conversely, the fertilized soil volume study was run for longer (90 days), but the volume of soil was smaller (500 g, approximately equivalent to soil in the 0 – 5 cm zone in the diffusion study), with a single sample representing the entire soil microcosm/pot. The larger soil volumes in the diffusion studies allowed the inhibitory effects of elevated pH, EC and aqueous NH$_3$ from standard urea to dissipate sooner, resulting in the more rapid commencement of significant rates of nitrification (Fig. 2a, b). The smaller volume of soil in the fertilized soil volume study clearly prevented dissipation of the effects of rapid urea hydrolysis (Fig. 6), and so, results are likely to overestimate the severity of nitrification inhibition in the standard urea treatments (Fig. 6). The fact that the PCU treatments still recorded significant amounts of nitrification in these restricted soil volumes (Fig. 6) emphasises the extent to which the impacts of urea hydrolysis were mitigated.

5 Conclusions

Polymer-coated urea bands mitigated the development of chemical conditions which inhibit nitrogen transformation processes in the fertosphere of standard urea bands. Consequently, nitrogen released from polymer-coated urea was rapidly nitrified, with this effect creating an extended window of risk for nitrogen losses from polymer-coated urea via denitrification and leaching loss pathways. As release of nitrogen from polymer-coated urea relies on diffusion from the granule into the soil solution, the close proximity of granules under banded (cf. mixed) applications further slowed nitrogen release by reducing concentration gradients between individual granules and the surrounding soil solution. Banding may therefore increase the complexity of matching nitrogen availability from controlled release fertilizers with plant demand. Furthermore, the delayed release of nitrogen from banded polymer-coated urea reduced the volume of nitrogen-enriched soil compared to that of urea bands, with potential implications for effective interception of applied nitrogen by plant roots—especially in soils and situations where hydraulic fluxes are limited. Soil characteristics influenced the rate of urea-nitrogen release from polymer-coated urea granules, possibly through differences in volumetric water content or processes associated with soil texture and carbon content. The present study highlights the importance of interactions between fertilizer application method, soil characteristics and moisture dynamics that collectively influence nitrogen release and soil cycling dynamics from polymer-coated urea. This study may provide preliminary insights for understanding processes that impact nitrogen availability in field conditions.

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Data Availability Supporting data is provided in supplementary material.

Declarations

Conflict of Interest The authors declare no competing interests.

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