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3	Decreasing ammonia loss from an Australian pasture with the use of enhanced efficiency			
4	fertilizers			
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6	Author names and affiliations			
7	Shu Kee Lam <sup>a*</sup> , Helen Suter <sup>a</sup> , Mei Bai <sup>a</sup> , Charlie Walker <sup>b</sup> , Arvin R. Mosier <sup>a</sup> , Hans van			
8	Grinsven <sup>c</sup> , Deli Chen <sup>a*</sup>			
9	<sup>a</sup> School of Agriculture and Food, Faculty of Veterinary and Agricultural Sciences, The			
10	University of Melbourne, Victoria 3010, Australia			
11	<sup>b</sup> Incitec Pivot Fertilisers, Seabreeze Parade, North Shore, Victoria 3214, Australia			
12	<sup>c</sup> PBL Netherlands Environmental Assessment Agency, The Hague, The Netherlands			
13				
14	*Corresponding authors:			
15	shukee.lam@unimelb.edu.au; delichen@unimelb.edu.au			
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#### 19 Abstract

Mitigating ammonia (NH<sub>3</sub>) volatilization from intensive pasture systems is critical for 20 environmental sustainability. However, field-scale evaluation on the potential of enhanced 21 22 efficiency fertilizers (e.g. urease inhibitors and controlled-release fertilizers) in mitigating NH<sub>3</sub> volatilization is limited. Using a micrometeorological technique, we conducted two field 23 trials to investigate the effects of Green UreaNV<sup>®</sup> (urea coated with the urease inhibitor *N*-(*n*-24 butyl)thiophosphoric triamide, NBPT) and polymer-coated urea (a controlled-release 25 fertilizer) on NH<sub>3</sub> volatilization from an intensive rainfed pasture in southern Australia. We 26 found that  $NH_3$  volatilization from urea was 5.8 and 5.6 kg N ha<sup>-1</sup>, respectively, in the 27 autumn and spring trials, equivalent to 11-12% of the applied urea in each season. The use of 28 Green UreaNV<sup>®</sup> and polymer-coated urea decreased the cumulative NH<sub>3</sub> volatilization by 29 45–55% and 80%, respectively. Taking into consideration the high environmental damage 30 cost of  $NH_3$  as found in the European Union, we hypothesize that both Green  $UreaNV^{$ <sup>®</sup> and 31 polymer-coated urea can be cost-effective in mitigating NH<sub>3</sub> loss from this pasture. Our 32 33 findings suggest that the extra cost of using these enhanced efficiency fertilizers for farmers is not compensated by the fertilizer N value of decreased NH<sub>3</sub> loss. However, from a societal 34 perspective the extra cost for Green UreaNV<sup>®</sup> is likely outweighed by reduced environmental 35 cost of NH<sub>3</sub>. New fertilizer technology should be developed to improve the cost-effectiveness 36 of polymer-coated urea to the farmers. 37

*Keywords*: ammonia volatilization, pasture, enhanced efficiency fertilizers, environmental
damage cost, sustainability

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## 41 **1. Introduction**

Ammonia (NH<sub>3</sub>) emission is conducive to the formation of fine secondary particulate matter
(PM<sub>2.5</sub>) in the atmosphere (Schiferl et al., 2014; Wu et al., 2016). When deposited, NH<sub>3</sub> can

cause eutrophication of aquatic and terrestrial ecosystems, and nitrous oxide (N<sub>2</sub>O) emission (Mosier et al., 1998; Galloway et al., 2008). The adverse impact of agricultural NH<sub>3</sub> emissions on human health and ecosystems has been estimated at  $\in$  10–120 billion per year in the European Union (van Grinsven et al., 2013) and updated to  $\in$  40–120 billion per year taking into account improved evidence for health impacts of NH<sub>3</sub> aerosols and impacts on marine ecosystems (van Grinsven et al., 2018). This highlights the urgency of NH<sub>3</sub> mitigation.

Ammonia mitigation measures include the use of enhanced efficiency fertilizers such as urea 51 52 granules coated with a urease inhibitor, and controlled-release fertilizers, which are proven to be effective when used in various agricultural systems (e.g. Pan et al., 2016; Xia et al., 2017). 53 54 Urease inhibitors are compounds that inhibit urease activity, thereby allowing the urea to 55 move into the soil before hydrolysis, and subsequently reduce NH<sub>3</sub> volatilization (Chen et al., 56 2008). Controlled-release fertilizers such as polymer-coated urea control the release of N from urea via the polymer coating which acts as a physical barrier, thus satisfying plant 57 58 requirements while maintaining low mineral N availability in the soil (Chen et al., 2008).

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Fertilizer N application in the Australian dairy industries increased rapidly from 21 kg N ha<sup>-1</sup> 60 in 1990 to 94 kg N ha<sup>-1</sup> in 2012 (Stott and Gourley, 2016); urea is the most commonly used N 61 fertilizer in pastures (Havilah et al., 2005). However, there are limited field-scale studies on 62 63 the quantification of NH<sub>3</sub> volatilization from Australian dairy pastures (e.g. Prasertsak et al., 2001; Eckard et al., 2003; Denmead et al., 2004), and very few on the potential of enhanced 64 efficiency fertilizers in mitigating this important pathway of N loss (Suter et al., 2013; Lam et 65 66 al., 2018). This is likely because a reliable quantification of NH<sub>3</sub> volatilization at the field scale requires the use of micrometeorological techniques (Denmead, 1983; Cichota and Snow, 67 2012), which involve huge labour and resource inputs, and large land area. In this study, we 68

69 conducted two field-scale measurements at a dairy pasture in a temperate region in southern 70 Australia using a micrometeorological method. The objectives of the study were to 71 investigate the effects of a urease inhibitor and a controlled-release coating on NH<sub>3</sub> 72 volatilization from urea fertilizer in an intensive pasture system under open-air conditions, 73 and to assess the economic and societal benefits of these enhanced efficiency fertilizers in 74 mitigating NH<sub>3</sub> volatilization.

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### 76 2. Materials and Methods

## 77 2.1 Experimental site

Field experiments were conducted on a temperate pasture dominated by perennial ryegrass 78 (Lolium perenne) at Wye (38°01' S, 140°53'E), South Australia, Australia (Fig. 1), in autumn 79 (6-26 May 2014) and spring (3-23 October 2014). The average minimum and maximum 80 81 temperatures during the experimental period in autumn were 10.0°C and 18.7°C, respectively, and 7.5°C and 21.2°C in spring. The rainfall during the same period was 28.6 mm and 12.8 82 83 mm in autumn and spring, respectively (Bureau of Meteorology, 2013). A three-dimensional 84 sonic anemometer (CSAT3, Campbell Scientific) was established placed on the field site to measure wind speed and wind direction during the experimental period. The soil at the site is 85 86 classified as a Tenosol (Isbell, 1996) with 78% sand, 18% silt and 4% clay. The surface soil (0–0.10 m) had a pH (CaCl<sub>2</sub>) of 4.9, and contained 4.9% organic C, 0.47% total N, 38.9 mg 87  $kg^{-1} NH_4^+$ -N and 28.8 mg  $kg^{-1} NO_3^-$  at commencement of the experiment. 88

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#### 90 2.2 Experimental design

Three treatments were included in this field experiment, viz. (i) urea (control), (ii) Green UreaNV<sup>®</sup> (urea coated with the urease inhibitor *N*-(*n*-butyl)thiophosphoric triamide, NBPT), and (iii) polymer-coated urea. Urea, Green UreaNV<sup>®</sup> and polymer-coated urea granules were

surface applied at a rate of 50 kg N ha<sup>-1</sup> on 6 May and 3 October to a 50 m diameter circular experimental area, with one circular area assigned to each treatment. The adjacent experimental areas were separated by at least 100 m to avoid cross-contamination of NH<sub>3</sub> (<u>Fig. 2</u>).

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# 2.3 NH<sub>3</sub> volatilization

A micrometeorological mass-balance method was used to determine the  $NH_3$  flux from each treatment (Freney et al., 1985). Two passive  $NH_3$  samplers (Leuning et al., 1985) were placed at 0.8 m above the ground surface on a mast located at the centre of each experimental area (Fig. 2). This height of 0.8 m is denoted as ZINST, or the stability independent height, which allows satisfactory determination of  $NH_3$  loss irrespective of the atmospheric stability (Wilson et al., 1982; Denmead, 1983).

Background measurements were made from the two sides of the experimental area along the 106 dominant wind direction, using two masts with the same height as the treatment circles. 107 108 These masts were located at least 100 m from the edge of the closest circle (Fig. 2). Following fertilizer application in the first week, NH<sub>3</sub> measurements were made twice daily 109 during the periods 0800–1700 h and 1700–0800 h (overnight). When the NH<sub>3</sub> flux gradually 110 decreased in the second week, measurements were made daily, and every two to three days 111 when the flux declined further in the third week. For each measurement, NH<sub>3</sub> captured in the 112 Leuning samplers was eluted with 40 mL MilliQ water and analyzed for NH<sub>4</sub><sup>+</sup>-N with a 113 segmented flow analyzer (SAN<sup>++</sup>, Skalar) (Keeney and Nelson, 1982). Ammonia flux for 114 each measurement period was calculated as described by Freney et al. (1985) and Leuning et 115 116 al. (1985), and summed over time to compute the cumulative emission.

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118 2.4 Soil N

119 Soil samples (0-5 cm) were collected with a 2.5-cm (internal diameter) auger from the circular treatment areas daily or every second day for a week after fertilizer application, then 120 every second or third day for the second week, and once in the final week. For each quarter of 121 122 the treatment circles 10 soil cores were sampled and composited. A subsample (10 g) was taken from the composited sample and kept frozen at -20°C until extraction with 100 mL 2M 123 KCl containing 10 mg  $L^{-1}$  phenylmercuric acetate. The extract was filtered with Whatman No. 124 42 filter papers and analyzed for urea and NH<sub>4</sub><sup>+</sup>-N on the Skalar SAN<sup>++</sup> segmented flow 125 analyzer. 126

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## 128 2.5 Statistical analysis

Data of soil urea and  $NH_4^+$  contents were analyzed with MINITAB 16 statistical package using ANOVA. The least significant difference (l.s.d.) at p = 0.05 was used to compare the means between the treatments (four replications). It was not possible to replicate  $NH_3$ measurements owing to the large area required for each treatment (~2,000 m<sup>2</sup>), which is common for large-scale  $NH_3$  flux measurements using the same micrometeorological technique (e.g. Turner et al., 2010). Nevertheless, this technique is robust for measurements of trace gas fluxes with uncertainty of within 15% (Denmead et al., 1998).

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### 137 **3. Results and discussion**

#### 138 *3.1 Seasonal variation of ammonia volatilization*

In the autumn trial,  $NH_3$  volatilization peaked three days after application of urea (9 May) and remained low (< 0.5 kg N ha<sup>-1</sup> day<sup>-1</sup>) thereafter for all treatments (Fig. <u>1a3a</u>). In contrast, NH<sub>3</sub> volatilization in the spring trial was steady and lasted for around 12 days after urea application (Fig. <u>1b3b</u>). Ammonia volatilization is dependent on edaphic and environmental factors such as soil pH, wind speed and soil moisture (Freney et al., 1983). The volatilization loss in our study site was driven by urea hydrolysis creating an alkaline hot spot (Du Preez and Burger,\_1988) in this otherwise acidic environment (soil pH of 4.9). The average wind speed during the autumn and spring trials was 2.8 and 4.0 m s<sup>-1</sup>, respectively. This is desirable for NH<sub>3</sub> volatilization because wind blows NH<sub>3</sub>-rich air away from the soil surface and hence increases the NH<sub>3</sub> gradient between the air and the soil surface, and subsequently the rate of volatilization (Bolan et al., 2004).

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In the autumn trial, the surface soil was sufficiently moistened by the heavy rainfall events 151 (~50 mm on 3–5 May) before urea application on 6 May. The light rainfall events (1–2 mm) 152 on 6 and 7 May allowed the urea granules to be dissolved but not diffused into the subsoil, 153 154 resulting in an abrupt increase in NH<sub>3</sub> volatilization on 9 May (Fig. <u>1a3a</u>). On 10 May the 7.6 155 mm rainfall was sufficient to wash the urea down in the soil, and since thenafter that low NH<sub>3</sub> emission was noted measured. This is supported by the decline in urea content from 6 May to 156 10 May (Fig. 2a4a). The effect of rainfall on NH<sub>3</sub> volatilization in our study is consistent with 157 the findings by Sanz-Cobena et al. (2011), who found that low water input (3 mm rainfall) 158 increased NH3 emission but the emission became minimal when urea was washed into the 159 160 subsoil by higher water input (7–14 mm rainfall).

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In the spring trial, no rainfall events were recorded for four consecutive days prior to urea application on 3 October, rendering the surface soil dry, which delayed urea hydrolysis and NH<sub>3</sub> volatilization (Black et al., 1987). The light rainfall events on 6–8 October (3.6 mm) and 13 October (2.6 mm) triggered NH<sub>3</sub> volatilization (Fig. <u>1b3b</u>). During these rainfall events the urea granules were gradually dissolved, as indicated by the increase in urea content from 6 October (Fig. <u>2b4b</u>). Ammonia volatilization became minimal after the 6 mm rainfall on 16 Oct, which likely washed all the urea down into the soil profile. The above results suggest that rainfall events played an important role in  $NH_3$  volatilization from this pasture, and that the timing of fertilizer application should be adjusted based on the likelihood of adequate rainfall events so as to minimize  $NH_3$  volatilization.

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## 173 3.2 Mitigation of ammonia volatilization by enhanced efficiency fertilizers

The cumulative  $NH_3$  emission for the urea treatment was 5.8 and 5.6 kg N ha<sup>-1</sup> over the 174 autumn and spring experimental periods, respectively, equivalent to 11–12% of the N applied. 175 This percentage of N loss is comparable to the global average of 16% loss of N as NH<sub>3</sub> from 176 177 surface applied urea in various agricultural systems (Pan et al., 2016). The N loss represents an economic loss to farmers of Australian dollars (AUD) 7.8  $ha^{-1}$  (5.7 kg N  $ha^{-1} \times 1.37$  AUD 178  $kg^{-1}$  N) (Table 1). More importantly, the potential damage cost of NH<sub>3</sub> to human health 179 (particulate matter) and ecosystem (eutrophication, biodiversity) in the European Union has 180 been estimated at € 10–30 per kg NH<sub>3</sub>-N (van Grinsven et al., 2013; 2018). Mitigation of 181 NH<sub>3</sub> volatilization is therefore urgently needed to improve N use efficiency and 182 environmental quality. 183

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Ammonia volatilization can be decreased by adopting various mitigation strategies, as 185 reported in recent global meta-analyses (Pan et al. 2016; Xia et al. 2017; Ti et al. 2019), 186 including the use of urease inhibitors and controlled-release fertilizers. In our study, NH<sub>3</sub> 187 emission in the autumn and spring trials was decreased by the Green UreaNV<sup>®</sup> (NBPT as the 188 urease inhibitor) to 2.6 and 3.1 kg N ha<sup>-1</sup>, respectively, representing a 45–55% reduction (Fig. 189 **<u>43</u>**). Likewise, the polymer-coated urea decreased NH<sub>3</sub> volatilization to 1.1 kg N ha<sup>-1</sup> in both 190 trials, equivalent to an 80% reduction (Fig. 13). The reduction induced by the urease inhibitor 191 and the polymer-coated urea is similar to the ranges (53-54 and 57-68%, respectively) 192 reported in the meta-analyses on NH<sub>3</sub> mitigation strategies (Pan et al., 2016; Ti et al., 2019). 193

194	The urease inhibitor NBPT inhibited urease activity and delayed urease hydrolysis (Sommer
195	et al., 2004; Chen et al., 2008), thereby decreasing NH <sub>3</sub> volatilization. This is shown by the
196	generally higher soil $NH_4^+$ content in the Green UreaNV <sup>®</sup> treatment than the urea treatment
197	in both trials (Fig. 5a, b). increase in The average soil $NH_4^+$ content increased from 107 (urea
198	treatment) to 152 mg kg <sup>-1</sup> (Green UreaNV <sup>®</sup> treatment) in the autumn trial ( $p < 0.05$ ), and
199	from 99 to 129 mg kg <sup>-1</sup> in spring ( $p < 0.05$ ). The relatively high soil NH <sub>4</sub> <sup>+</sup> content (> 100 mg
200	$kg^{-1}$ ) in this pasture was likely because $NH_4^+$ was retained in the soil, rather than being
201	volatilized, in this acidic soil environment, combined with the relatively high background N
202	due to past management of the pasture site. The polymer-coated urea, as a controlled-release
203	fertilizer, slowed down-urea release from the fertilizer granules (Chen et al., 2008).
204	Consequently, throughout the experimental period, the $NH_4^+$ content under the treatment of
205	polymer-coated urea (averaging 44 and 64 mg kg <sup><math>-1</math></sup> in autumn and spring, respectively) was
206	much lower than the urea treatment ( $p < 0.05$ ) (Fig. 5a, b), and NH <sub>3</sub> volatilization was also
207	decreased.

It is worth noting that fertilizers with urease inhibitor and controlled-release coatings are not 209 widely adopted despite their effectiveness in mitigating NH<sub>3</sub> volatilization from various 210 agricultural systems globally. One major obstacle preventing the wider adoption is that the 211 extra cost of these fertilizer products (Table 1) may not secure productivity benefits (e.g. 212 Suter et al., 2013; Nauer et al., 2018), as also observed in this field site (Suter et al., 213 unpublished). This lack of productivity response in pastures has been attributed to non-214 limiting background soil N, unfavourable climatic conditions and differential responses of 215 pasture species (Suter et al. 2013; Di and Cameron, 2016). We found that there were no 216 financial benefits to farmers because the fertilizer N value of NH<sub>3</sub> saved by using Green 217 UreaNV<sup>®</sup> and polymer-coated urea was lower than the extra cost of these products (Table 1). 218

This gap could be narrowed <u>if these products can be produced at a lower cost or if subsidies</u>
are provided to farmers for using the products.

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222 Nonetheless, a far more important but often ignored concern is the environmental sustainability of the system. As mentioned, the estimated environmental damage cost of NH<sub>3</sub> 223 is € 10–30 per kg NH<sub>3</sub>-N (van Grinsven et al., 2013). We performed a simple economic 224 assessment for this Australian pasture based on the increase in fertilizer cost and the decrease 225 in environmental damage cost of NH<sub>3</sub> associated with the use of Green UreaNV<sup>®</sup> and 226 polymer-coated urea, when compared to urea (Table 1). We found the environmental damage 227 cost (NH<sub>3</sub>) avoided by using Green UreaNV<sup>®</sup> (AUD 46–137 ha<sup>-1</sup>) would outweigh the 228 additional cost of this fertilizer product (AUD 6.4 ha<sup>-1</sup>) compared to urea. This results in net 229 societal benefits of AUD 39–130  $ha^{-1}$  (Table 1). While the damage cost of NH<sub>3</sub> loss has not 230 been estimated for Australia, it is likely lower than that of the EU because of the lower NH<sub>3</sub> 231 emission density and higher proportion of urban population in Australia than in the European 232 countries, resulting in a stronger spatial disconnection between potentially exposed 233 population and NH<sub>3</sub> emission sources (Crippa et al., 2018; Liang et al., 2018; Population 234 Reference Bureau, 2018). Therefore, the lower bound of our estimates on societal benefits is 235 more realistic for Australia. In contrast, when the environmental damage cost of NH<sub>3</sub> is low, 236 the use of polymer-coated urea would not be cost-effective at the current market price in 237 decreasing NH<sub>3</sub> loss (Table 1). Its cost-effectiveness will improve with the increase in the 238 environmental damage cost of NH<sub>3</sub> (Table 1). This underpins the need of a cost-effective 239 coating for controlled-release fertilizers. 240

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In summary, we recommend that the environmental damage cost of N loss associated with fertilizer use should be considered when evaluating the mitigation potential of enhanced

efficiency fertilizers, and that this potential can be enhanced with the advancement in fertilizer technology. The potential societal benefits of enhanced efficiency fertilizers provide solid ground for subsidizing these products to compensate farmers for additional costs, particularly in regions with higher risk of NH<sub>3</sub> exposure of humans and ecosystems. <u>This</u> would promote the wider adoption of these products in agriculture for environmental <u>sustainability.</u>

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367	Figure captions						
368	<u>Fig. 1.</u>	Location of experimental site (Wye)					
369							
370	<b>Fig. 2.</b>	Aerial layout of the experimental site for NH <sub>3</sub> measurement by two passive					
371		samplers mounted on a mast (					
372		with urea (blue circle), Green UreaNV <sup>®</sup> (green circle), and polymer-coated					
373		urea (red circle), and the background locations (not drawn to scale)					
374							
375	Fig. <mark>13</mark> .	Cumulative ammonia (NH <sub>3</sub> ) volatilization from the application of urea, Green					
376	UreaNV <sup>®</sup> , and polymer-coated urea in the autumn (a) and spring (b) trials						
377							
378	Fig. <mark>24</mark> .	Urea content in the soil under the treatment of urea, Green $UreaNV^{\text{(B)}}$ , and					
379	polymer-coated urea in the autumn (a) and spring (b) trials. Error bars represent standard						
380	errors of the mean of four replicates.						
381							
382	Fig. 5	$NH_4^+$ content in the soil under the treatment of urea, Green UreaNV <sup>®</sup> , and					
383	polymer-coate	ed urea in the autumn (a) and spring (b) trials. Error bars represent standard					
384	errors of the n	nean of four replicates.					

# Tables Click here to download Tables: Table 1\_final.docx

# Table 1

Extra fertilizer cost, reduction of  $NH_3$  loss, and net benefits of using Green Urea $NV^{\text{®}}$  and polymer-coated urea, taking into consideration of the environmental damage cost of  $NH_3$ 

	Urea	Green	polymer-
	(control)	UreaNV®	coated urea
Fertilizer cost (AUD $t^{-1}$ ) <sup>a</sup>	630	689	1500
Fertilizer N cost (AUD kg <sup>-1</sup> N) <sup>b</sup>	1.37	1.50	3.26
Fertilizer application at 50 kg N $ha^{-1}$			
Fertilizer N cost at (AUD $ha^{-1}$ )	68.5	74.9	163.0
Extra fertilizer cost relative to urea (AUD $ha^{-1}$ ) (I)		6.4	94.6
Average cumulative NH <sub>3</sub> loss of the two trials (kg N $ha^{-1}$ )	5.7	2.85	1.1
Amount of NH <sub>3</sub> saved relative to urea (kg N ha <sup><math>-1</math></sup> )		2.85	4.6
Fertilizer N value of $NH_3$ saved relative to urea (AUD ha <sup>-1</sup> ) (II)		3.9	6.3
Benefit to farmers (AUD $ha^{-1}$ ) (II – I)		-2.5	-88.3
Amount of $NH_3$ damage cost avoided relative to urea (AUD ha <sup>-1</sup> ) (III)		46 to 137	74 to 221
Societal benefit relative to urea (AUD $ha^{-1}$ ) (III – I) <sup>c</sup>		39 to 130	-21 to 126
<sup>a</sup> Insites Divet Fontilisons nort price list (affective Fohmery 2010)			

<sup>a</sup> Incitec Pivot Fertilisers port price list (effective February 2019)

<sup>b</sup> calculated based on the content of N in urea (46%)

<sup>c</sup> Environmental damage cost of NH<sub>3</sub>, estimated to be  $\in$  10–30 per kg NH<sub>3</sub>-N (van Grinsven et al., 2013; 2018), was converted into Australian dollars (AUD) 16–48 per kg NH<sub>3</sub>-N based on exchange rate of AUD: Euro = 1.6:1.



Fig. 1

Figure 2 Click here to download Figure: Figure 2.docx



Fig. 2



Fig. 3







