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Title: Assessment and mitigation of the environmental burdens to air from land applied food-based digestate

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Abstract (limit 150 words only)

Anaerobic digestion (AD) of putrescible urban waste for energy recovery has seen rapid growth over recent years. In order to ascertain its systems scale sustainability, however, determination of the environmental fate of the large volume of digestate generated during the process is indispensable. This paper evaluates the environmental burdens to air associated with land applied food-based digestate in terms of primary pollutants (ammonia, nitrogen dioxide) and greenhouse gases (methane and nitrous oxide). The assessments have been made in two stages – first, the emissions from surface application of food-based digestate are quantified for the business as usual (BAU). In the next step, environmental burden minimisation potentials for the following three mitigation measures are estimated - mixed waste digestate (MWD), soil-incorporated digestate (SID), and post-methanated digestate (PMD). Overall, the mitigation scenarios demonstrated considerable NH₃, CH₄ and N₂O burden minimisation potentials, with positive implications for both climate change and urban pollution.

Keywords: *anaerobic digestion; bio fertilizer; digestate; environmental burdens; OFMSW*

Capsule abstract: In situ monitoring and analyses demonstrate the role of post-processing in greenhouse gases and air pollution mitigation from food-based digestate use as bio fertiliser.

Highlights:

- In situ air pollution assessment of land applied digestate is performed.
- Environmental burden minimisation scenarios for digestate bio fertiliser presented.
- Food-based digestate show high ammonia volatilisation potential.
- Soil incorporated digestate effectively reduces NH₃ but elevates N₂O emissions.
- Managing digestate emissions mitigate both climate change and air pollution.

1 **1. Introduction**

2 Digestate, the semi-solid residue obtained post biogas extraction in anaerobic digestion (AD), is considered
3 a vital source of organic matter and nutrients, especially nitrogen. It is increasingly being applied as soil
4 conditioner/amendment (on urban gardens, farmlands, recreation/sports grounds, fish ponds, etc.), or
5 alternatively for developing energy crops on brownfield/marginal land and sports turf production (WRAP,
6 2013). This is mainly due to its two attributes - one, for providing a low carbon substitute for fossil fertilisers
7 (Chambers and Taylor, 2013; WRAP, 2011); two, for restoring soil organic matter and for closed-loop
8 nutrient recycling, especially mineral nitrogen (Fricke et al., 2007; Karagiannidis and Perkoulidis, 2009;
9 Möller and Stinner, 2009). Consequently, digestate field application has been identified as a sustainable
10 practice in terms of meeting the EU standards for good agricultural and environmental condition (GAEC)
11 (RPA/Defra, 2012). However, with greater emphasis on strategies for diverting biowastes from landfill and
12 their sustainable re-utilisation through valorisation in AD, the volumes of digestate are expected to increase
13 rapidly (typical digestate represents 70-95% of the feedstock volume) (Lukehurst et al., 2010). Digested
14 slurries have been found to be significant sources of ammonia (NH₃), methane (CH₄) and nitrous oxide
15 (N₂O) emissions (Amon et al., 2006; Bacenetti et al., 2013; Nkoa, 2014; Wulf et al., 2002a), with potential
16 implications for local-to-regional climate (NRC, 2002; Ravishankara et al., 2009) and human health (Peel
17 et al., 2013). Nkoa's (2014) detailed review has suggested that digestates can be considered as organic
18 amendments (or organic fertilizers) only when properly handled and managed.

19
20 The content and quality of digestate depends largely on both the feedstock and the hydraulic retention time
21 (HRT) of the digester; usually longer HRT reduces the organic content owing to more effective
22 methanogenesis (Szűcs et al., 2006). Digestate quality is further affected by maturing in storage tanks
23 (Menardo et al., 2011). Rigid compliance criteria for Class I digestate have been set by the European
24 Commission (EC) and the British Standard Institution (BSI) (BSI, 2010; EC, 2014). Although the scale of
25 AD operation dedicated to organic waste treatment is at an all-time high (and on the rise), there is still
26 relatively little published information on the composition and potential environmental behaviour of
27 digestate, particularly from AD plants processing food wastes (Tiwary et al., 2015; Whelan et al., 2010).
28 Anaerobic digestion of slurry tends to increase ammoniacal-N concentrations, reduce carbon to nitrogen
29 ratios and increase pH, thereby increasing the risk of NH₃ losses during storage and soil application (Möller
30 and Stinner, 2009). Inappropriate disposal of unstable digestate on land may lead to formation of residual
31 biogas, with potential health issues from exposure to its constituent non-methanic volatile organic
32 compounds and other hazardous air pollutants and odorous compounds (Palmiotto et al., 2014).

33

34 During AD, the majority of organic (slow release) nitrogen is transformed into readily available nitrogen
35 (RAN) (Defra, 2011), specifically for protein-rich feedstocks, including the organic fraction of the
36 municipal solid waste (OFMSW)¹, dairy by-products and slaughterhouse waste (Kryvoruchko et al., 2009;
37 Menardo et al., 2011). Typical total-N content of food-based digestate ranges between 5-8 kg m⁻³, with
38 about 60-80% of this present as RAN; the proportion of RAN to total-N in food-based digestate is nearly
39 40% higher than manure-based digestate (202 kg and 145 kg respectively for every 250 kg total-N) (WRAP,
40 2011). Moreover, for digestate from kitchen waste feedstock RAN of as much as 99% of corresponding
41 Total-N has been reported (Furukawa and Hasegawa, 2006). Sensitivity analyses conducted to assess the
42 split share of digestate total-N (i.e. organic-N, NH₄-N and NO₃-N) on acidification potential have reported
43 NH₃ and NO₃ as the main contributors to the enrichment of pollutants in direct air and water environments
44 respectively (Bernstad and la Cour Jansen, 2011; Evangelisti et al., 2014). Free ammonia concentration is
45 affected mainly by temperature, pH and total ammoniacal nitrogen (TAN) concentration (Chambers and
46 Taylor, 2013). Whereas a number of multivariate data analysis protocols using advanced sensor
47 technologies have become available, the majority of these are currently (2014) limited to AD process
48 monitoring; there are limited approaches for digestate quality monitoring and they are all based on offline
49 instrumentation (Oppong et al., 2012). Minimum quality requirements for whole digestate, separated liquor
50 and separated fibre have been prescribed as part of a Publicly Available Specification (PAS 110)² standards
51 in the UK (BSI, 2010). PAS 110 Clause 10 provides guidance on obtaining representative samples of all
52 three types of digestates (whole, fibre, liquor) via one or more sampling access points appropriately located
53 in the digestate production/storage system prior to its use.

54

55 The knowledgebase involving environmental impacts from digestate land application is largely developed
56 from traditional approaches for cattle manure management, which requires incorporating the unique and
57 emerging characteristics of OFMSW digestate. From an environmental point of view there is an existing
58 challenge of reducing the gaseous N-losses (NH₃, NO₂, N₂O) from digestate soil application to the fields
59 (Balsari et al., 2007; Misselbrook et al., 2005a; Nyord et al., 2008; Tiwary et al., 2015). This study aims to
60 determine and interpret the seasonal emissions profile to air from digestate slurry applied to urban soil. The
61 latter has been assessed through small plot experiments, applying digestate acquired from community-scale
62 AD operations. It evaluates the implications for climate change and urban pollution in terms of fitness for
63 purpose of the available practice, including some proposed mitigation strategies. Preliminary outputs from
64 field trials of in situ monitoring of soil-applied digestate for air emissions of N-pollutants (NH₃, NO₂) and

¹ OFMSW is defined by the European Commission as “biodegradable park and garden waste, food and kitchen waste from household, restaurants, caterers and retail premises and comparable waste from food processing plants”.

² PAS (Publicly Available Specification) is a sponsored fast-track standard driven by the needs of the client organisations in the United Kingdom, developed according to guidelines set out by British Standard Institute.

65 greenhouse gases (GHGs: CH₄, N₂O) are reported. These results feed into a review of approaches to
66 mitigate the environmental burdens while promoting re-utilisation potentials of anaerobically digested
67 slurry as marketable bio fertilisers.

68

69

70 **2. Materials and methods**

71 *2.1. Review of sampling and analytical techniques*

72 A review of available techniques was deemed essential for designing a robust assessment methodology for
73 evaluating the emissions from both the baseline and the responses to plausible mitigation strategies.

74

75 *2.1.1 Primary air pollutants*

76 Several studies have reported monitoring of NH₃ emissions from land-applied manure in Europe (Gericke
77 et al., 2011; Loubet et al., 1999; Misselbrook et al., 2005a; Nyord et al., 2012; Nyord et al., 2008), Australia
78 (Leuning et al., 1985), USA (Parker et al., 2013) and Canada (King et al., 2012). Inter comparison of
79 different monitoring techniques are also reported (NRC, 2002; Parker et al., 2013) (Misselbrook et al.,
80 2005b). The following three broad categories of approaches have been applied for estimating air pollutant
81 emission rates from area sources: i) Micrometeorological mass balance techniques, where fluxes are
82 calculated from a large footprint using tower-based instrumentation (e.g., gradient methods, eddy
83 covariance); ii) Indirect methods, where ambient concentrations are measured and source emission rates
84 are either back-calculated using dispersion models or using known transfer factors from standards (also
85 known as ‘standard comparison’ method; iii) Direct methods, where fluxes are measured from samples
86 collected from the source using portable wind tunnels or flux chambers, followed by colorimetric/FTIR
87 spectroscopy. Among these, micrometeorological mass balance methods have been found to provide more
88 realistic estimates, thus widely used in field monitoring (Generemont et al., 1998; Nyord et al., 2012;
89 Wilson and Shum, 1992), but limited to applications over larger source areas with sufficient fetch for
90 integration of emission rates. Standard comparison methods are popular owing to their cost-effectiveness
91 (Möller and Stinner, 2009; Wulf et al., 2002b) but are quite sensitive to surface characteristics and
92 meteorological conditions, requiring regular corrections to the fluctuations in the transfer factor applied in
93 emissions estimates. In particular, such methods assume the transfer factors estimated from standard plots
94 to be valid for calculating the emission rates from the experimental plots, overlooking the inconsistencies
95 in micrometeorological conditions, especially the wind fields between the standard and the experimental
96 plots. Despite their limitations, portable wind tunnels and flux chambers are often the only available, direct
97 method for assessing air pollutant fluxes from small individual area sources. Theoretical principles of wind
98 tunnel technique have been described in detail in the literature (Loubet et al., 1999; Misselbrook et al.,

99 2005b). The appropriateness and accuracy of portable wind tunnels and flux chambers for quantifying area
100 source emissions have been extensively discussed in the literature (Fowler et al., 2001; King et al., 2012;
101 Loubet et al., 1999; Misselbrook et al., 2005b; Nyord et al., 2012; Parker et al., 2013).

102
103 Qualitative assessment of the cost-benefits of the different monitoring techniques have also been reported
104 (see for example (Gericke et al., 2011; Misselbrook et al., 2005b)). For small-plot comparative
105 measurements, a wind tunnel system is recommended, albeit with a caveat of its tendency to modify the
106 temperature at the emitting surface relative to the ambient conditions owing to canopy effect (Braschkat et
107 al., 1993; Misselbrook et al., 2005b; Nyord et al., 2012), thereby influencing the emission rates. Application
108 of wind tunnel technique is thus far best suited for making comparative measurements for treatments
109 imposed on small plots and less representative for estimating the absolute air emissions from large area
110 sources.

111
112 2.1.2. Greenhouse gases

113 Compared to monitoring of primary air pollutants, there appears to be a greater consistency in the reported
114 literature on monitoring of greenhouse gases (GHG) from area sources. The majority of the studies have
115 applied closed chambers (static or mobile) for sampling of N₂O and CH₄, followed by concentration
116 estimations using gas chromatography (Amon et al., 2006; Wulf et al., 2002a). With closed chambers,
117 however, reportedly there is an issue with negative feedback of increasing gas concentration in the chamber
118 on the rate of diffusion of the gases, implying that the rate of release is often very high immediately after
119 land application.

120
121
122 2.2. *Sampling and analysis*

123 2.2.1 Experimental design

124 A dedicated wind tunnel experimental set up was developed following Lockyer (1984) and Loubet et al.
125 (1999), comprising of an upwind-downwind gas sampling system and an air extractor (fitted with an
126 anemometer to monitor the average wind speeds through the tunnel canopy). The front portion of the wind
127 tunnel was fabricated using transparent polycarbonate material covering 1 m² (2 m long x 0.5 m wide),
128 offering exposure to direct sunlight and ensuring negligible temperature interference from the tunnel to the
129 treated surface area. Three separate wind tunnels were operated simultaneously in parallel (**Fig. 1**) to
130 acquire statistically representative samples (Misselbrook et al., 2005a; Nyord et al., 2012). Air was drawn
131 from the upwind/downwind sampling ports located on top of the tunnel into impinger tubes containing the
132 absorbing media through three sets of polytetrafluoroethylene (PTFE) tubing (approximately 4 mm internal

133 diameter) at a flow rate of 3 l min^{-1} using an arrangement of two manifolds and suction pumps. For GHG,
134 only the downwind port of the middle tunnel was used to draw the sample from an air-tight chamber inserted
135 into the soil to a depth of 10 cm.

136

137 2.2.2 Field sampling

138 All land application experiments were conducted at a field site representative of tropical climates, located at
139 The Energy and Resources Institute (TERI) campus (suburbs of New Delhi, 28.45°N , 77.03°E), India.
140 Three wind tunnels, each covering an exposed area of 1 m^2 , were operated in parallel on a $4 \text{ m} \times 4 \text{ m}$ plot
141 (**Fig. 1**), and predominantly composed of sandy loam soil ($\text{pH} = 5.5$, organic matter content (%) = 3.7). The
142 wind tunnels were positioned such that the canopy inlets were located at the upwind edge of the plots and
143 they remained in the same position for the entire duration of each experiment. To avoid cross-interference
144 of emissions between the neighbouring wind tunnels care was taken to restrict the spread of fresh digestate
145 slurry within the exposed portion of each tunnel using a watering can. The amount of digestate spread on
146 different plots was volume-limited to 0.02 m^3 (i.e. 20 L), with a spread area of 1 m^2 and application
147 thickness of 2 cm per plot. All the experiments were conducted with no precipitation recorded within the
148 first 24 h following digestate application.

149

150 Air pollutants (NH_3 , NO_2) were sampled over 6 consecutive days following digestate application based on
151 the literature (Nyord et al., 2012; WRAP, 2011). Although previous experiments using digested cattle slurry
152 have reported NH_3 emissions to cease after 48 h (Amon et al., 2006), a relatively longer sampling period
153 was chosen for food-based digestate, potent to exhibit longer emission patterns owing to higher RAN
154 (Chambers and Taylor, 2013; Whelan et al., 2010). A quality assurance run was performed to establish the
155 sampling protocols for NH_3 using two-serially connected impinger tubes to the downwind port. This
156 experiment showed that less than 2% of the total NH_3 trapped during the sampling reached the second
157 impinger tube, which confirmed the emissions were insufficient to fully saturate the absorbing media in the
158 first impinger tube. Therefore, for all subsequent experiments only one impinger tube was used for the
159 entire sampling. During each experiment, the impinger tubes filled with absorbing media, and connected to
160 the upwind/downwind sampling ports of all the three wind tunnels, were simultaneously exposed
161 continuously at a stretch for 4 h (corresponding to sampling medians of 2, 26, 50, 74, 98 and 122 h).

162

163 Monitoring of greenhouse gases (CH_4 , N_2O) was performed from an air-tight chamber connected to the
164 downwind sampling port of the middle wind tunnel owing to logistical limitation in simultaneous operation
165 of gas chromatograph. Following land application, CH_4 was monitored for up to 7 days on a daily basis
166 (Wulf et al., 2002a); N_2O was monitored for up to 30 days at a sampling interval of 0, 1, 2, 3, 5, 9, 16, 23

167 and 30 days, with reduced sampling frequency beyond the first week as per the literature (Amon et al.,
168 2006; Wulf et al., 2002a).

169

170 2.2.3 Analysis

171 Methodological details of the analysis adopted for different digestate parameters (dry matter (DM) content,
172 pH, total Kjeldahl N (TKN) content, total ammoniacal N (TAN) content, etc.) are provided in **Table 1**.
173 Analyses of primary air pollutants followed the standard practice for quantifying ambient air concentrations
174 by bubbling a known volume of air through the impinger tubes filled with absorbing media and connected
175 to the upwind/downwind sampling ports of the tunnels. NH₃ was quantified through the Indophenol method
176 (Method 401, Air Sampling and Analysis, 3rd Edition, (CPCB, 2011)) using a dilute solution of sulphuric
177 acid as absorbent to precipitate the airborne ammonia as ammonium sulphate followed by its colorimetric
178 analysis by reaction with phenol and alkaline sodium hypochlorite to produce indophenol. NO₂ was
179 quantified through the modified Jacobs & Hochheiser method (IS5182 Part 6, Methods for Measurement
180 of Air pollution: Oxides of nitrogen (CPCB, 2011)) by bubbling the sample through a solution of sodium
181 hydroxide and sodium arsenite to convert the airborne NO₂ into nitrite ion (NO₂⁻¹). In the subsequent step,
182 this was colorimetrically analysed by reacting with phosphoric acid, sulphanilamide and N-(1-naphthyl)-
183 ethylenediamine di-hydrochloride (NEDA). The emission per sampling period was calculated as the
184 product of the volume of air passing through the tunnel and the difference in outlet and inlet air
185 concentrations. For each measuring period, the background pollutant concentration from the inlet located
186 on the upwind face of the tunnel was estimated and subtracted from the cumulative concentration estimated
187 from the port located on the downwind face of the exposed area.

188

189 The GHGs were analysed by means of a gas chromatograph (GC-5700 series, Nucon Engineers, New Delhi)
190 using a 1.8 m (~6 ft) long Poropak-Q column with thermal conductivity detector (TCD). The injector port,
191 the detector and the oven were operated at temperatures of 40, 40 and 35°C respectively. Argon was used
192 as a carrier gas at a pressure of 1.8 kg cm⁻². One ml of gas sample was injected using a micro syringe into
193 the gas chromatograph for analysis.

194

195

196 2.3 Scenario analysis

197 The following scenarios were developed and tested sequentially using the experimental facility. Details of
198 the physico-chemical properties of the digestate used in this analysis are presented in **Table 2**. For primary
199 air pollutant monitoring (NH₃, NO₂), the cumulative emissions for the different scenarios were estimated
200 as arithmetic means of three replicates (**Fig. 1**), as it is considered more robust (and less biased) for small

201 numbers of replicates (Wulf et al., 2002a). The overall emission trends were obtained by fitting regression
202 curves to the experimental data in SigmaPlot v12.5 (Systat Software Inc.).

203

204 2.3.1 Business as usual

205 As a first step, assessments were conducted for the baseline emission patterns (BAU) following land
206 application of digestate obtained from processing 100% OFMSW in a two-stage AD plant, available at the
207 study site. The digestate was relatively fresh when land applied, keeping the approach consistent with the
208 growing emphasis on reducing the storage time of digestate used as soil amendments, in order to avoid
209 nutrient (mainly RAN) and carbon loss.

210

211 2.3.2 Mixed-waste digestate

212 Typically, mixed-waste feedstock (with higher C/N ratio) have tendency to improve biogas yield and lower
213 NH₃ release potentials from digestate. Co-digestion of OFMSW with cattle manure is commonly adopted
214 in wet digestion (Banks et al., 2011). On the other hand, co-digestion of waste having high N-content (food
215 waste, vegetable waste, food processing industry waste and slaughterhouse waste) with waste paper (typical
216 mix of 95% to 5% respectively) is proposed for controlled dry digestion (Li et al., 2011; Takata et al., 2013),
217 owing to its collateral benefits of adjusting the C/N ratio of the medium and regulating the accumulation of
218 both NH₃ and VFA in the reactor. As the first mitigation measure, performance of a mixed waste digestate
219 (MWD), obtained following co-digestion of 70% OFMSW (food, fruit and vegetable waste) with 30% cattle
220 dung, was evaluated to optimise the earmarked emissions.

221

222 2.3.3 Soil incorporated digestate

223 Uniform application of digestate slurry near or under the soil surface is recommended as an environmental
224 best practice in the literature (Amon et al., 2006; Nyord et al., 2012). However, ‘sub-surface incorporation’,
225 has demonstrated mixed results in abatement of gaseous emissions. While it positively mitigates NH₃
226 emissions (Chambers and Taylor, 2013; Nyord et al., 2012; Wulf et al., 2002b), it tends to exacerbate the
227 GHG emissions under anaerobic conditions (CH₄, N₂O) (Möller and Stinner, 2009; Nkoa, 2014; Wulf et
228 al., 2002a). The proposed mitigation measure of soil incorporated digestate (SID) involved shallow
229 injection (5-7 cm below ground) of the BAU digestate, followed by its immediate soil incorporation based
230 on the best available technique (BAT) recommended in the literature (Brizio and Genon, 2010).

231

232 2.3.4 Post-methanated digestate

233 Typically during the first 2 months of digestate storage post-methanation, up to 15% additional CH₄ yields
234 have been reported (Balsari et al., 2013; Menardo et al., 2011; Weiland, 2010). However, digestate storage

235 during post-methanation also have collateral influence on NH₃ emissions, owing to high ammonium
236 nitrogen (NH₄-N) concentration (Whelan et al., 2010). Previously reported NH₃ emission rates ranged
237 between 2.06 and 4.44 g NH₃ m⁻² and between 7.89 and 14.6 g NH₃ m⁻² from stored, whole digestate and
238 from digested liquid fraction respectively (Gioelli et al., 2009). The post-methanated digestate (PMD) for
239 this study was obtained after maturing the BAU digestate into a 150 L concrete tank with a floating dome
240 for 45 days.

241
242

243 **3 Results and discussion**

244 The sampling and analyses steps described above were repeated to obtain the corresponding emissions for
245 NH₃, NO₂, CH₄ and N₂O in order to evaluate the environmental performance of the proposed mitigation
246 scenarios MWD, SID and PMD with reference to the BAU. The following sections describe the observed
247 trends as well as their seasonal variations, if any; these are discussed in the context of developing effective
248 digestate handling and management strategies to achieve reduced environmental burdens and enhanced
249 economic values, the latter in terms of improved nutrients and organic matter reutilisation.

250
251
252

253 *3.1 Emissions trends*

254 3.1.1 Ammonia emissions

255 The NH₃ emissions for all the scenarios showed a common trend of highest emissions within the first two
256 days and negligible emissions beyond the four-day threshold (**Fig. 2a**), which agrees with previous studies
257 (Amon et al., 2006; Möller and Stinner, 2009; Wulf et al., 2002a). Preliminary results suggest that digestate
258 surface application leads to significant losses of NH₃ to air over a short span during the first week, with the
259 business as usual having the highest cumulative NH₃ emissions following land application, estimated to be
260 over 65% of the applied TAN (**Fig. 2b**). Compared to this, application of mixed feedstock digestate and
261 post-methanated digestate had cumulative emissions around 45% and 35% of applied TAN respectively,
262 resulting in over 35% NH₃ reductions over the business as usual from these interventions. However, the
263 maximum NH₃ abatement of around 85% was achieved from slurry soil incorporation immediately
264 following application; the cumulative emissions estimated in this case was around 10% of the applied TAN
265 and negligible NH₃ emissions were observed from the second day onward (< 1% of TAN), which was along
266 the lines of previous reportings (Nyord et al., 2012; Wulf et al., 2002a).

267
268

269 3.1.2 Nitrogen dioxide emissions

270 The majority of AD literature has extensively reported on NH₃ emissions whereas NO₂ emissions from
271 digestate application are not adequately accounted for in the emissions inventories. NO₂ emissions from
272 soil amended plots are considered highly uncertain, and strongly influenced by the soil microenvironment
273 (MNP, 2007); estimates from a previous digestate assessment study suggest it to be up to 15% of the NH₃
274 emissions in the Netherlands (de Vries et al., 2003). Results from our study showed overall trends for NO₂
275 emissions similar to NH₃, with BAU having the largest cumulative emissions, followed by MWD while
276 SID showed negligible NO₂ emissions (**Fig. 3**). The rate of emissions for both BAU and MWD were very
277 feeble past 24 h from land treatment, and their respective cumulative emissions were estimated within 10%
278 and 4% of TAN.

279

280 3.1.3 Methane emissions

281 Compared to NH₃ the observed CH₄ emission intensity was noted to be relatively short-lived for BAU, with
282 a steep decline in emissions from day 3 onwards (**Fig. 4**). Nevertheless, based on our analysis, in the initial
283 1-2 days from land application, BAU had the highest CH₄ emissions intensity, followed by MWD. PMD,
284 despite showing similar emissions trend temporally, had a much reduced intensity (up to 56% lower)
285 compared to the BAU. This is mainly attributed to residual CH₄ extraction from post-methantion; reported
286 estimates of residual CH₄ potentials during digestate maturing/long term storage (up to 180 day) vary,
287 ranging from 5-15% to 12-31% of total methane production (Weiland, 2003). These variations are linked
288 to the feed quality, the organic loading rate (OLR) and the HRT of the AD process, as well as the moisture
289 content of the digestate itself, typically reported residual CH₄ potential for animal manure, energy crops
290 and food industry waste range between 2.88 and 37.63 L kg⁻¹ volatile solids (Menardo et al., 2011). The
291 steep hike in emissions in the initial phase (within 1-2 days) post-application for BAU, MWD and PMD is
292 mainly attributed to the readily available dissolved CH₄, produced during storage of the substrate. On
293 contrary, SID showed a lower emission intensity in the first 24 hour, peaking only after 2 days of land
294 application. These trends of delayed peaking of CH₄ from injected slurry is also found in previous
295 monitoring campaigns (Wulf et al., 2002a) and can be mainly attributed to kick-starting of sub-surface
296 anaerobic degradation of VFAs through methanogenesis under humid conditions.

297

298 3.1.4 Nitrous oxide emissions

299 Nitrous oxide is formed as an intermediate product of both nitrification and de-nitrification. Previous studies
300 have reported strong increase in N₂O emissions from mitigation strategies for reducing NH₃ volatilisation
301 involving either application of liquid digestate, typically with a narrow C/N ratio and high soil infiltration
302 levels (Möller and Stinner, 2009), or injection and soil incorporation of digestate (Wulf et al., 2002a).

303 Indirect N₂O production from emitted NH₃ has been considered a potential indirect contributor to global
304 warming (Wulf et al., 2002a), with approximately 1% of NH₃-N assumed to be re-emitted to the atmosphere
305 as N₂O-N (IPCC, 2001). However, this mechanism was negated as a possible source for N₂O in our
306 experiments and all N₂O was attributed to direct emissions, given the use of wind tunnel and the timescale
307 of samples collected. The BAU and MWD showed similar levels of emissions, PMD had relatively lower
308 emissions but SID showed a peculiar, and quite contrasting emission trend, with increasing levels of
309 emissions recorded about two weeks post land application (**Fig. 5**), which is also reported in previous
310 studies (Möller and Stinner, 2009; Nyord et al., 2008). The background N₂O emissions from the bare soil
311 have also been shown in this figure for reference.

312

313

314 *3.2. Seasonal effects*

315 There is evidence of increased air emissions following digestate soil application on environmental factors,
316 such as ambient temperature, wind speed and precipitation (Chambers and Taylor, 2013; Nyord et al., 2012;
317 Parker et al., 2013; Peel et al., 2013). This section reports outcomes of the air emissions evaluated for two
318 contrasting periods: summer (August – October) and winter (January – March) (**Table 3**). Based on the
319 outputs from the monitoring experiments, potential environmental burdens for NH₃, NO₂, CH₄ and N₂O to
320 air were estimated over the two seasons as a function of the fresh matter digestate mass applied to soil for
321 the BAU, MWD and SID scenarios (**Table 4**). Distinct seasonal characteristics influencing the emissions
322 have been identified below.

323

324 Sunlight hours - Our results showed strong dependence of the monitored emissions on sunlight hours; the
325 observed trends as NH₃ >> N₂O > CH₄, whereas NO₂ showed an inverse dependence. For all the trace gases
326 the cumulative emissions remained unaltered and the effect of cooler and moist periods during winter
327 mainly impeded their release rates, as reported in previous studies (Wulf et al., 2002a). NO₂ emissions were
328 marked with large fluctuations and were found to be higher during overcast winter months (typically with
329 average ambient temperature around 10°C and less than 2 hours of direct sunlight on the field plots).

330

331 Wind speed - In previous studies, wind speed has been reported as one of the parameters with the greatest
332 influence on NH₃ emissions from slurries (Misselbrook et al., 2005a). As followed in this study, the wind
333 tunnel set up reportedly provides a more robust wind-sensitive NH₃ emissions estimates (Rong et al., 2009).
334 The contrasting wind micro-environment recorded over the two seasons, with up to 75% reduction in winter
335 over summer (**Table 3**), seems to have resulted in large differences between the observed emissions,
336 especially for NH₃.

337 Soil moisture - Lower wind speed during winter resulted in sustained build-up of humid conditions inside
338 the tunnels, enhancing the soil moisture content (**Table 3**). This in turn resulted in poor soil respiration,
339 contributing to reduced emissions during this period. On contrary, NH₃ losses were intensified during
340 summer when slurries were applied to dry soils under warm weather conditions. For CH₄, the dry conditions
341 facilitated formation of crusts on whole digestate, resulting in possibility of some anaerobic release of
342 residual CH₄. Also, in case of SID, the increased soil temperature triggered sub-soil anaerobic processes,
343 resulting in relatively higher N₂O emissions over summer.

344

345

346 **4 Conclusions and future directions**

347 This study demonstrates application of simultaneous *in situ* monitoring of air pollutants (NH₃, NO₂) and
348 GHGs (CH₄, N₂O) following land application of digestate as bio fertiliser. The baseline emissions from the
349 business as usual (BAU) have been evaluated in the first step, followed by assessment of the corresponding
350 environmental burden minimisation potentials of three proposed mitigation measures – mixed waste
351 digestate (MWD); soil-incorporated digestate (SID); post-methanated digestate (PMD). Our results show
352 the proposed mitigation measures to be effective over BAU: NH₃ emissions are considerably reduced from
353 MWD and PMD (by up to 35% and 43% respectively) and significantly reduced from SID (by up to 85%).
354 However, delayed elevation peak for N₂O in case of SID, primarily attributed to sub-surface denitrification,
355 showed marginal increase in emissions over BAU (by up to 2%). On the other hand, PMD effectively
356 reduced CH₄ emissions (by up to 55%) with inconsiderable influence on other emissions. Nonetheless, we
357 acknowledge these outcomes as specific to food-based digestate; the effectiveness of the proposed
358 mitigation strategies to digestates arising from other feedstocks will vary, primarily owing to their different
359 physico-chemical characteristics. It is also noteworthy that the results are based on digestate characteristics
360 from our pilot-scale anaerobic reactor, which may differ for other digestate characteristics depending on
361 the type of substrate and the type of AD process adopted.

362

363 The study highlights some paradoxical future sustainability concerns for the voluminous amounts of
364 digestate bound to be inadvertently generated from increased processing of OFMSW using AD. It warrants
365 timely intervention for developing adequate strategy for post-AD handling and management of digestate,
366 ideally as a marketable product (e.g. bio fertiliser) to the urban allotment/vegetable gardeners and to the
367 wider farming communities. In addition, it solicits consideration for more advanced methods of digestate
368 processing and reutilisation, including - dewatering, storage, composting, curing, exploiting alternative
369 applications in construction/regeneration activities, etc.

370

371 Further, development of a practical software tool quantifying the emissions from digestate soil applications
372 would facilitate sustainable digestate management practice on a routine basis. In this respect, our
373 preliminary work on meteorological consideration can serve as useful stepping stones. It is noteworthy this
374 study reported baseline emissions from bare plots, assuming the soil amendments are meant to prepare the
375 land for cropping. Although some literature reports on the long term emission trends, incorporating the
376 responses from planted vegetation (see for example, Moller and Stinner 2009), these are primarily for rural
377 setting, and a more comprehensive study on the usage of digestate as soil amendment in the urban context
378 is recommended as a natural next step study in order to develop full appreciation of the coupled soil-
379 vegetation effects.

380

381

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Figure 5. Plot showing temporal trends in the differential nitrous oxide emissions to air for the four scenarios (expressed as microgram N). Shown alongside is the background emission from bare soil for reference.

Table 1.

Parameter	Method/Approach	Literature source
Dry Matter (% FM)	Oven drying at 105 °C	(APHA, 2005)
Organic Matter (%)		
pH	Digital pH meter (accuracy of ± 0.01 pH unit)	(APHA, 2005)
TKN in digestate (g (kg FM) ⁻¹)	Digestion of sample with H ₂ SO ₄ and use of Kjeldahl apparatus for distillation	(APHA, 2005)
TAN in digestate (g (kg FM) ⁻¹)	Distillation method	(APHA, 2005)
CH ₄ , N ₂ O in gaseous samples	Gas Chromatograph (NUCON 5700) equipped with auto sampler and a thermal conductivity detector and 6 feet long Poropak Q stainless steel column	(APHA, 2005)
NH ₃ , NO ₂ in gaseous samples ($\mu\text{g m}^{-3}$)	Shimadzu spectrophotometer (UV1700)	(CPCB, 2011; Misselbrook et al., 2005b)

TKN = Total Kjeldahl nitrogen; TAN = Total available nitrogen (as ammonia)

Table 2.

	BAU	MWD	PMD
pH	8.5	8.2	8.7
Dry matter (% solids v./v.)	4.3	5.6	3.2
Organic matter (%)	18.5	16.2	15.7
Total N (g)	147.2	120.6	128.7
RAN (g)	118.8	87.8	106.5
Total S (SO ₃ , g)	8.8	16.5	8.7
Specific gravity (kg m ⁻³)	0.99	0.98	0.99
Fresh matter (g)	19.8	19.6	7.3
Digestate volume (L)	20	20	20

(Note: BAU - 100% OFMSW; MWD - 70% OFMSW+30% cattle dung, v./v.; PMD – matured BAU)

Table 3.

	Avg. Air Temp. [z=5cm] (°C)	Avg. Soil Temp. [z=-5cm] (°C)	Avg. Wind Speed [z=25cm] (m s⁻¹)	Soil moisture (%)	Precipitation(mm)	Qualitative comments
<i>Summer experiments (Aug – Oct 2013)</i>	39	31	2	10	0.8	dry/sunny
<i>Winter experiments (Jan – Mar 2014)</i>	11	7	0.5	45	5	humid/ foggy

Table 4.

Feedstock type	NH ₃		NO ₂		CH ₄		N ₂ O	
	<i>S</i>	<i>W</i>	<i>S</i>	<i>W</i>	<i>S</i>	<i>W</i>	<i>S</i>	<i>W</i>
BAU	5.05	4.25	0.75	0.90	1.3E-02	1.0E-02	8.5E-04	4.2E-04
MWD	2.69	1.55	0.30	0.45	1.1E-02	7.0E-03	6.3E-04	3.5E-04
SID	0.79	0.27	0.04	0.05	1.2E-02	9.0E-03	8.6E-04	5.7E-04

S=summer (~40°C, dry/sunny); W=winter (~10°C, humid/foggy)

Fig. 1.

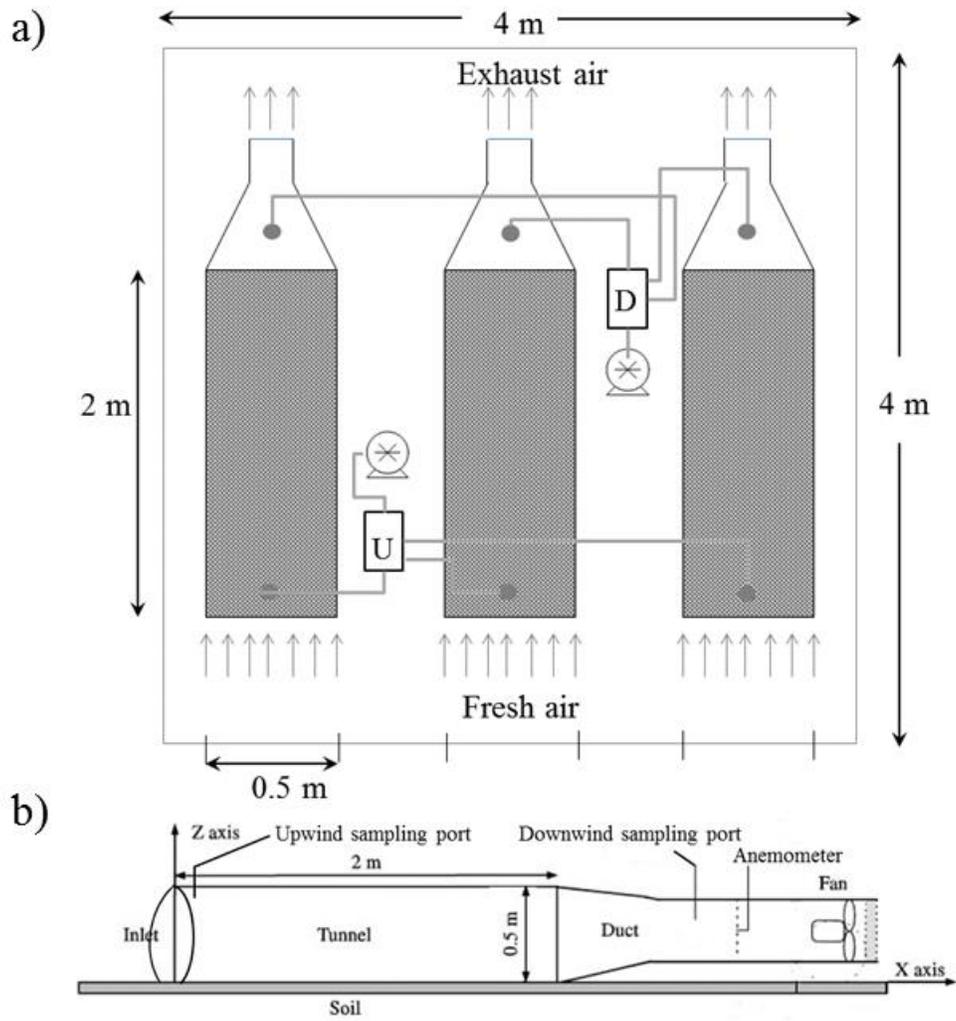
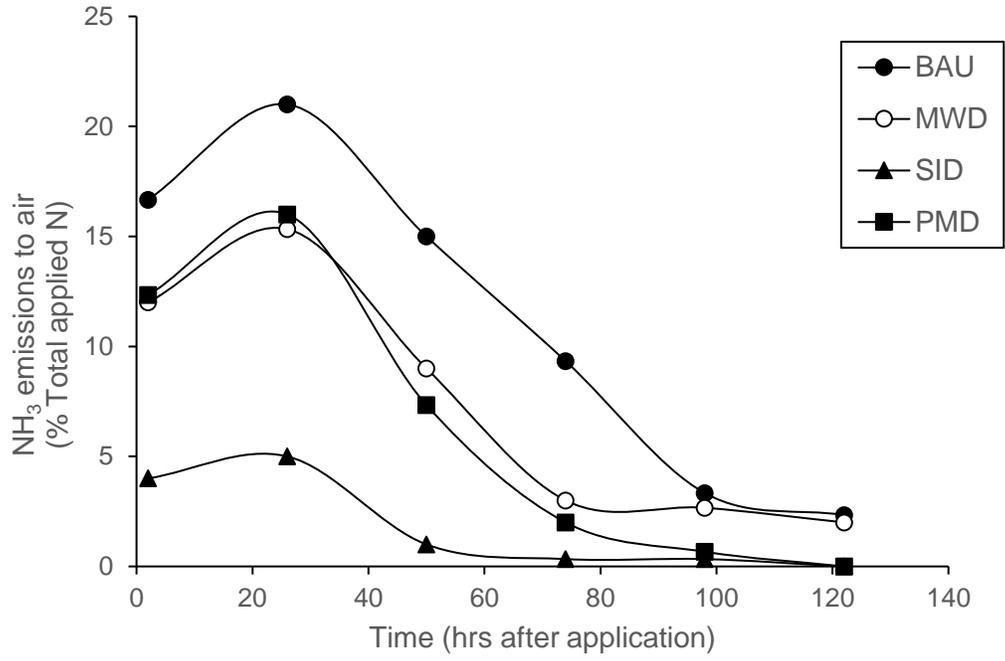


Fig. 2.

a)



b)

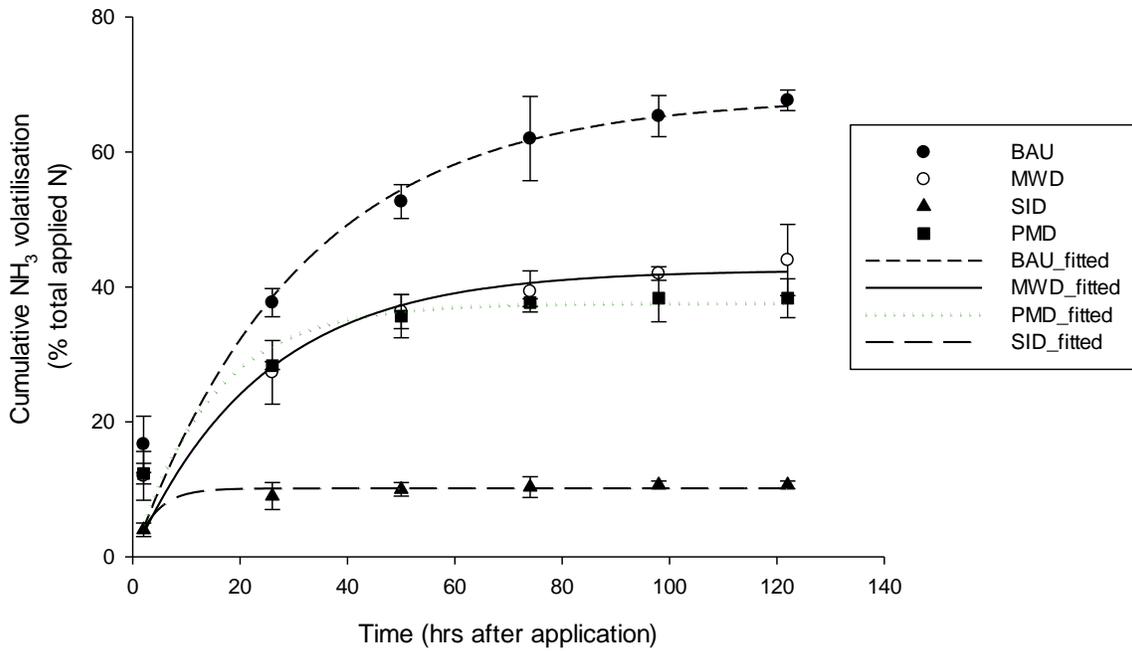


Fig. 3

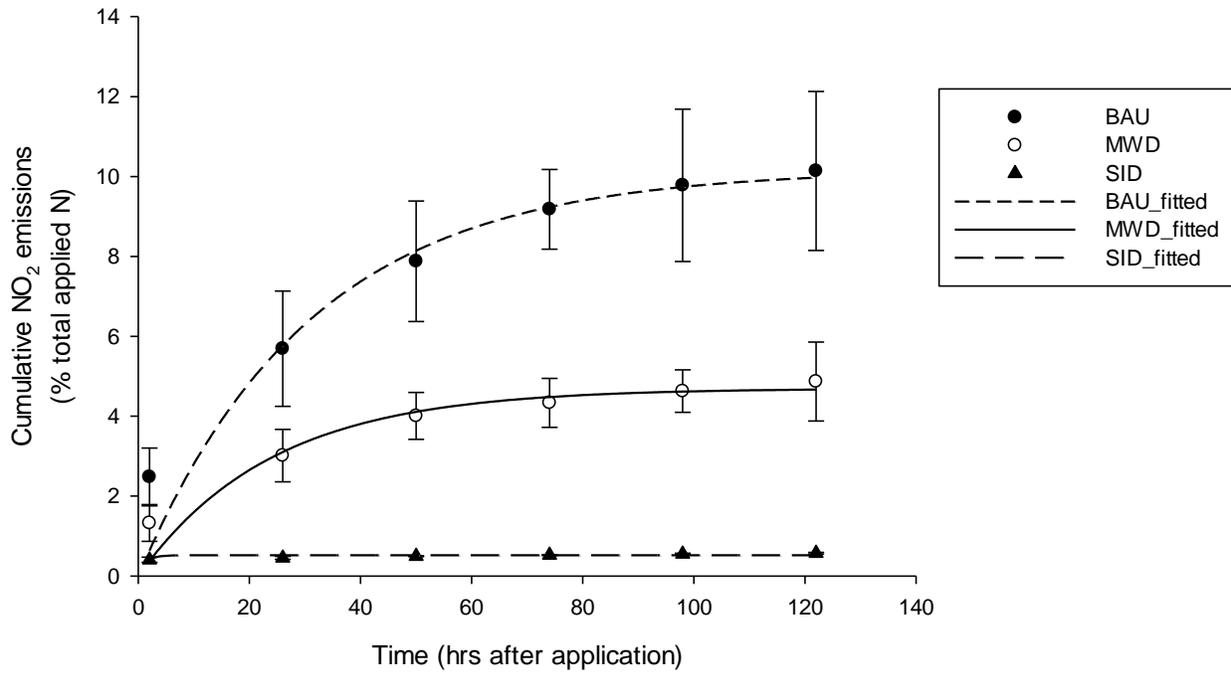


Fig. 4

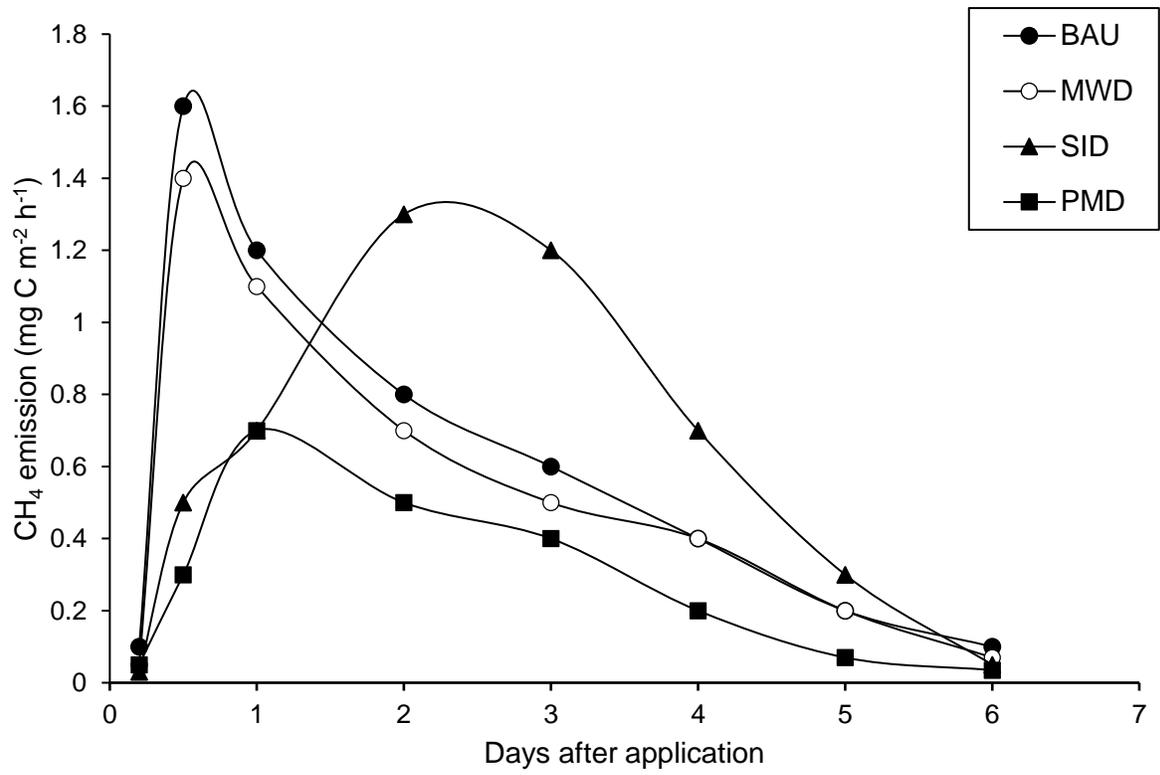


Fig. 5

