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1 Review paper intended for publication in *Waste Management*

2

3 **Trace Gas Emissions from Municipal Solid Waste Landfills: A**
4 **Review**

5

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7

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15

16 **Highlights:**

- 17 • Uncovered waste surfaces are the most important emission sources for trace gases.
18 • Landfills with non-source-separated waste and poor management give high emissions.
19 • Geomembranes/biocovers are more efficient in reducing fugitive trace gas emissions.
20 • Trace gases pose risks to human health and the environment in some countries.

21

22 **Keywords:** Origin and fate; landfill gas constituents and concentrations; sampling and
23 analysis; quantitative emission measurement; health risk; environmental impacts.

24

25 **Abstract**

26 Trace gas emissions from municipal solid waste (MSW) landfills have received increasing
27 attention in recent years. This paper reviews literature published between 1983 and 2019,
28 focusing on (i) the origin and fate of trace gas in MSW landfills, (ii) sampling and analytical
29 techniques, (iii) quantitative emission measurement techniques, (iv) concentration and surface
30 emission rates of common trace compounds at different landfill units and (v) the
31 environmental and health concerns associated with trace gas emissions from MSW landfills.
32 Trace gases can be produced from waste degradation, direct volatilisation of chemicals in
33 waste products or from conversions/reactions between other compounds. Different chemical
34 groups dominate the different decomposition stages. In general, organic sulphur compounds
35 and oxygenated compounds are connected with fresh waste, while abundant hydrogen
36 sulphide, aromatics and aliphatic hydrocarbons are usually found during the methane
37 fermentation stage. Selection of different sampling, analytical and emission rate measurement
38 techniques might generate different results when quantifying trace gas emission from landfills,
39 and validation tests are needed to evaluate the reliability of current methods. The
40 concentrations of trace gases and their surface emission rates vary largely from site to site,
41 and fresh waste dumping areas and uncovered waste surfaces are the most important fugitive
42 emission sources. The adverse effects of trace gas emission are not fully understood, and
43 more emission data are required in future studies to assess quantitatively their environmental
44 impacts as well as health risks.

45 **1. Introduction**

46

47 As a result of global economic development, population growth and urbanisation, more
48 and more municipal solid waste (MSW) is generated each year around the world. According
49 to the World Bank, 2.01 billion tonnes of MSW was generated in 2016, and up to 3.40 billion
50 tonnes will be generated by 2050 under a business-as-usual scenario (Kaza et al., 2018),
51 which will pose great challenges for governments. Among the various waste treatment and
52 disposal methods, landfilling is the leading method in most countries, especially in
53 low-income and upper-middle-income countries (Kaza et al., 2018). On a global level, about
54 70% of all generated MSW is disposed in some sort of landfill (sanitary and unsanitary
55 landfills, open dumps) (Fischedick et al., 2014; Kaza et al., 2018). Whereas landfilling type
56 (either controlled or uncontrolled) as well as disposed waste composition differ from country
57 to country, short- and long-term gaseous emissions – alongside associated human health and
58 environmental risks – represent one of the biggest concerns. Raw landfill gas (LFG) or source
59 gas, normally consisting of 50-60% methane and 40-50% carbon dioxide, is generated from
60 the biodegradation of disposed organic matter inside the landfill. In addition, LFG also
61 contains hundreds of different compounds in trace amounts (combined ~1% in the total
62 volume and referred to as ‘trace gases’), originating either from disposed waste products and
63 hazardous waste and/or from waste degradation processes occurring in the landfill body (US
64 EPA, 1997; Young and Parker, 1983).

65 Although trace gas emissions represent only a very small portion of the overall emission
66 from landfills, they continue to be a concern, due to the adverse effects on the environment
67 and human health. Compounds, such as benzene and vinyl chloride, are carcinogenic to
68 humans, and it is also widely recognised that sulphur compounds, together with some
69 oxygenated compounds and hydrocarbons, are responsible for the offensive odours emanating
70 from landfills. Moreover, emissions of chlorofluorocarbons and halocarbons contribute to
71 ozone depletion and global warming (Licata and Minott, 1996), and some alkanes and
72 aromatics combine with nitrogen oxides in the atmosphere to form secondary air pollutants
73 (Canzano et al., 2010).

74 In the 1970s and 1980s, research on trace gas emissions from MSW landfills was carried
75 out in the United States and some European countries (Parker et al., 2002; Shen et al., 1990).
76 The topics included trace gas composition and concentrations in raw LFG and in ambient air
77 from distinct locations at a landfill site, trace gas sampling and analytical techniques,
78 emission rate measurements, odour and health risk assessments, etc. Landfill gas composition
79 and trace gas concentrations were extensively explored and reported. Emission rates as well
80 as emission factors (in $\mu\text{g s}^{-1}$ or $\mu\text{g m}^{-2} \text{min}^{-1}$) for a range of trace gases from MSW landfills
81 were established by the United States Environmental Protection Agency (US EPA) (e.g. the
82 EPA AP-42 default values) (US EPA, 1995a; 1995b; 1997). Research published before 2002
83 was reviewed by Brosseau and Heitz (1994), Hamideh Soltani-Ahmadi (2000) and Parker et
84 al. (2002).

85 In the late 1990s, a sharp decline in trace gas emissions was observed from MSW
86 landfills in the US compared to the EPA AP-42 default values, mainly due to the
87 implementation of the US EPA's Resource Conservation and Recovery Act's (RCRA)
88 hazardous regulations leading to reduced amounts of hazardous waste entering these sites
89 (Hamideh Soltani-Ahmadi, 2000; Staley et al., 2006). Similarly, the European Union released
90 the Landfill Directive (CEC, 1999) in 1999, which has since contributed to a steady decrease
91 in biodegradable waste going into landfills and at the time was also expected to reduce trace
92 gas emissions significantly (European Environment Agency, 2016; Pawlowska et al., 2008).
93 Hence, data reported before 2000 do not represent the current status, and there is a need to
94 update knowledge on trace gas emissions from landfills in those countries. In recent years,
95 there has been rising concern in developing and middle-income countries, such as in China,
96 about the potential risks caused by trace gas emissions from MSW landfills. The lack of
97 source segregation, and the high organic content of landfilled waste as well as improper site
98 operations in some countries, has exacerbated trace gas emissions, potentially causing excess
99 health risks to surrounding populations (Liu et al., 2016a; Tian et al., 2013).

100 An increasing number of studies on trace gas emissions from MSW landfills have been
101 published within the last 10 years, and large amounts of data on trace gas composition,
102 concentrations and their associated environmental and health risks have been reported

103 globally (Fig. 1). However, most of the data focused on trace gas concentrations measured at
104 different landfill units, which are difficult to compare with each other, and trace gas emission
105 rates from MSW landfills were rarely reported. In addition, these studies have focused on
106 different compounds, making it challenging to understand the dominant chemical groups on a
107 general level. Hence, a new review paper is in need, to provide an overview of the current
108 trace gas emission status at MSW landfills, to update and compare data from studies in both
109 developed and developing countries, to provide insights into related environmental impacts
110 and health risk assessments and to identify future research needs in this field.

111

112 *****Fig.1.*****

113

114 This review article includes six sections based on studies published in 2003-2019. It
115 discusses the origin and fate of trace gases in MSW landfills, before moving on to introduce
116 and discuss the sampling and analytical techniques as well as quantitative emission rate
117 measurement techniques. Concentrations of some common trace compounds in raw LFG and
118 in ambient air at different landfill units are reported and compiled, and fugitive emission rates
119 of trace gases from previous studies are summarised and compared for different landfill
120 surfaces. In addition, a review on the adverse impacts of trace gas emission is undertaken,
121 concentrating on factors such as human health risks, odour pollution, stratospheric ozone
122 depletion, climate change and air pollution. Finally, research needs are identified for future
123 studies on trace gas emissions from MSW landfills.

124

125 **2. Definition, origin, transportation and emission of trace gases in MSW landfills**

126 **2.1 Definition of landfill trace gases**

127 Landfill trace gases are gaseous compounds present in trace amounts (a total volume
128 fraction of up to 1%) in raw LFG. In previous studies, trace components present in raw LFG
129 were named ‘non-methane organic compounds’ (NMOCs), ‘non-methane hydrocarbons’
130 (NMHCs), ‘volatile organic compounds’ (VOCs), ‘volatile compounds’ (VCs) or ‘odorous

131 compounds' (OCs), depending on the target compounds and the topic. In this paper, we use
132 the term 'trace gases', which mainly refers to VOCs but also includes inorganic compounds
133 such as hydrogen sulphide (H₂S) and ammonia (NH₃), and we divide them into seven
134 chemical groups according to their chemical structure and functional group (Dincer et al.,
135 2006; Keller, 1988; Komilis et al., 2004; Parker et al., 2002; Staley et al., 2006). Table 1
136 provides an overview of the seven defined chemical groups in this paper (a list of focusing
137 compounds can be found in the Supplementary Material).

138

139 *****Table 1*****

140

141

142 **2.2 Origin of trace gases in MSW landfills**

143

144 A conceptual figure, showing the generation, conversion, transportation and emission
145 processes of trace gases in MSW landfills, is provided as Fig. 2. Trace gases in MSW landfills
146 originate from three processes: 1) biodegradation of organic matter in disposed waste, which
147 produces biogenic-originated compounds, 2) direct volatilisation of compounds (often
148 xenobiotics) contained in waste products and/or chemicals disposed of at the landfill and 3)
149 chemical/biological conversation/reaction between different compounds or by-products.

150

151 *****Fig. 2.*****

152

153 Once waste goes into a landfill, it undergoes microbial degradation, which changes from
154 aerobic to anaerobic degradation over a certain period, depending on the landfill operation.
155 The decomposition of organic matter in the waste starts when it is collected, transferred and
156 transported, and conditions in the waste are mainly aerobic (Statheropoulos et al., 2005; Tan
157 et al., 2017; Zhao et al., 2015). This decay continues after the waste is tipped into the landfill,
158 and it lasts until the waste is compacted and covered and oxygen is totally consumed (i.e. the

159 initial adjustment stage and the transition stage). Thereafter, the anaerobic biodegradation of
160 the disposed organic matter starts, and it goes through three successive stages before
161 stabilisation is reached: acidogenesis, acetogenesis and methanogenesis stages (Christensen
162 and Kjeldsen, 1995). Fig. 3 shows a qualitative schematic view of trace gas abundance in raw
163 LFG during the different degradation stages. In the initial adjustment and transition stage, an
164 emission peak for all trace gases is often observed, which occurs mainly due to volatilisation
165 (Thomas and Barlaz, 1999). As the waste degradation process continues and the oxygen
166 content in the waste decreases, new biogenic-originated trace gases are produced, whereas the
167 emission of most xenobiotic compounds decreases. When the waste enters the
168 methanogenesis stage, another concentration peak of all trace compounds in LFG might be
169 observed, due to the production of large amounts of by-products and intermediates from
170 anaerobic biodegradation together with the gas-stripping effect (Canzano et al., 2010; Chiriac
171 et al., 2011; Davoli et al., 2003; Moreno et al., 2014; Staley et al., 2006; Thomas and Barlaz,
172 1999). The concentrations of all trace gases in LFG continue to decrease along with the
173 stabilisation of the landfill until they reach close to zero. A more detailed discussion on the
174 development of each chemical group over time will be provided in Section 5 herein.

175

176 *****Fig. 3.*****

177

178 **2.3 Trace gas transportation and emission**

179 After being generated inside the waste body, trace gases can go through various
180 processes before they are collected or emitted into the environment. Important processes
181 include diffusion (driven by concentration differences), advection (driven by pressure
182 differences), dilution, volatilisation, adsorption, conversion and degradation, depending on the
183 physiochemical properties of the individual trace gas (Abdul-Wahab et al., 2016; Reinhart,
184 1993). Fig. 2 shows the transportation and emission processes of trace gases in an MSW
185 landfill.

186 After waste is disposed of at a landfill, some trace gases are emitted directly into the

187 atmosphere during the dumping, compression and compaction processes, and this emission
188 will continue for a certain period, if the waste is exposed to ambient air without cover. When
189 the waste is covered, a proportion of the trace compounds produced or released from the
190 waste accumulates in the gas phase, and eventually it is extracted and destroyed in LFG flares
191 or engines at sites with gas collection and recovery. Uncollected LFG and trace gases
192 contained therein rise to the landfill surface through diffusion and advection and are then
193 emitted into the atmosphere (i.e. fugitive emissions). During transportation to the landfill
194 surface, the adsorption of trace gases to waste materials and conversion/degradation in the
195 waste body/soil cover might happen (Tassi et al., 2009). For landfills without gas extraction
196 systems, or with passive LFG collection systems, pressure might build up inside the landfill,
197 which will enhance the advective and diffusive transportation of trace gases , resulting in
198 significant fugitive emissions of trace gases (Kjeldsen, 1996; USEPA, 1995; Xie et al., 2016).
199 Water-soluble trace compounds might dissolve into infiltrating water and move directly into
200 the leachate collection system (at controlled landfills), which then may possibly be released
201 into the atmosphere at the leachate storage/treatment units through evaporation (Kjeldsen and
202 Christensen, 2001; Shen et al., 1990). At uncontrolled landfills without leachate collection,
203 trace gases that have dissolved into the leachate might leach into groundwater. In addition,
204 trace compounds contained in leachate can also transport through bottom liner systems at
205 considerable rates (Edil, 2003).

206 The generation of trace gases is mainly governed by the original waste composition and
207 the degradation stage of the waste. Nevertheless, their concentrations in LFG and in ambient
208 air as well as emission rates from landfill surfaces, is governed by both production and
209 transportation processes (USEPA, 1995), and is affected by many factors, including
210 pre-treatment of waste, landfill management operations, waste compaction status, climatic
211 conditions, the type of LFG collection and cover systems, etc. , , (Capanema et al., 2014;
212 Chiemchaisri et al., 2010; Pawlowska et al., 2008; Sadowska-Rociek et al., 2009; Urase et al.,
213 2008). For example, sorting out of organic waste fraction could effectively reduce the
214 production of some odorous trace gases (Lou et al., 2015; Zhang et al., 2012a), while
215 compaction activity on-site usually lead to increased release of trace compounds at the

216 working surface (Chiriac et al., 2007). The climatic condition (dry or wet) together with waste
217 compaction status (density) and the cover geotechnical properties can also affect trace gas
218 emissions. In wet season, the pore spaces inside waste/cover material are occupied by water,
219 which would limit the transport of gases and reduce the available sorption sites for trace gases
220 on the waste/cover material. In dry seasons, an opposite situation is developed, and the
221 eventual surface emission is dependent on which process is the controlling mechanism
222 (Yesiller et al., 2018). In addition, under dry conditions cracks might develop in the cover (e.g.
223 on clay cover) leading to increased advective flow of LFG from the landfill surface (Scheutz
224 et al., 2008). Installation of active LFG collection systems reduces the pressure accumulation
225 inside the landfill body and leading to a reduced advective force, which could effectively
226 decrease trace gas emissions from the surface. However, active gas extraction may allow air
227 ingress to landfills, resulting in incomplete aerobic conditions where the waste will enter
228 initial acidogenesis and acetogenesis stages and large amount of odorous oxygenated
229 compounds are produced (Parker et al., 2002). Different cover types could also significantly
230 affect trace gas emission from the landfill, and this will be discussed more in detail in section
231 5 and 6.

232 Important landfill units that might be responsible for trace gas emissions include the
233 active waste dumping area (working face), covered surface (temporary and final covers),
234 leachate collection, storage or treatment pool, leakage points in the LFG collection system
235 and cracks in surface covers, LFG flares, etc. The concentrations of trace gases measured in
236 ambient air at different landfill units and their surface emission rates will be discussed in
237 section 5 and section 6.

238

239 **3. Sampling and analytical techniques**

240

241 Proper sampling is fundamental for measuring landfill trace gas concentrations and
242 quantifying emission rates. The sampling procedure must be carefully designed to ensure that
243 representative samples are taken and properly stored, thus avoiding any loss or alteration in
244 the composition and concentration so that they reflect the real state of the monitored air or gas.

245 Precise analytical techniques are also necessary, especially when considering the low
246 concentration levels and diverse chemical composition of trace gases. In fact, the number of
247 compounds that can be analysed in a study is often restricted and determined by the sampling
248 and analytical techniques adopted, which could easily affect the final conclusions. When
249 conducting a study on landfill trace gases or making comparison to previous studies, one
250 should always take into account the specific methodologies applied to avoid possible bias in
251 the conclusions.

252 Various techniques exist for collecting trace gas samples from different types of emission
253 sources, and these can be classified as whole-air sampling techniques and solid sorbent
254 trapping techniques. There are also many highly developed analytical techniques with
255 different instrumentation for analysing trace gas samples, including various ex-situ and
256 on-site monitoring technologies. Review papers have been published on the sampling,
257 pre-concentration and analytical techniques of VOCs in environmental air samples (Hamideh
258 Soltani-Ahmadi, 2000; Król et al., 2010a, 2010b; Ras et al., 2009). In addition, the US
259 Environmental Protection Agency (US EPA) has introduced a series of standardised
260 procedures for determining VOCs in ambient air by using vacuum canisters, Tedlar bags,
261 sorbent tubes and gas chromatography (GC) or gas chromatography-mass spectrometry
262 (GC-MS), e.g. TO-14, TO-15, TO-17 (US EPA, 1999a, 1999b, 1999c). This section presents a
263 brief review of trace gas sample collection and analysis at MSW landfills. The advantages and
264 limitations of selected commonly used sampling techniques, as well as their applications, are
265 summarised in Table 2. Emission rate measurement techniques will be introduced separately
266 in Section 4.

267

268 ***3.1. Sampling techniques***

269 *3.1.1. Whole-air sampling techniques*

270 Whole-air sampling, as the name suggests, involves collecting gaseous samples in
271 different containers – without losing any constituents in the original air sample. The major
272 advantage of whole-air sampling is that it is easy to perform, and large volumes of air or gas

273 can be sampled within a short time frame. Instantaneous as well as time-weighted
274 concentrations can be provided, depending on the sampling time and flow rate. In addition,
275 multiple gas samples can be extracted for analysis from the obtained sample, which
276 minimises analytical errors (Hamideh Soltani-Ahmadi, 2000). Containers that are widely used
277 for whole-air sampling include sampling bags made from different synthetic materials,
278 vacuum canisters and glass-bombs, which are discussed in detail below.

279 **Sampling bags.** Sampling bags made from different plastic materials are the most
280 widely used containers for trace gas sampling at landfills. According to EN 13725 of the
281 European Committee for Standardisation (CEN, 2003), sampling bags can be made from one
282 of the following inert materials: tetrafluoroethylene hexafluoropropylene copolymer (FEP),
283 polyvinyl fluoride (PVF, Tedlar) or polyethylene terephthalate (PET, Nalophan). Air sampling
284 into plastic bags is often performed using the ‘lung method’, i.e. the bag is placed into an
285 air-tight container connected to a vacuum pump. When the pump starts to work, the air inside
286 the container is removed and thus pressure drops, causing the bag to fill with the targeted air
287 (Fig. 4). This method avoids direct contact between the sampled air and the pump, thus
288 avoiding possible contamination from the latter.

289 **Canisters.** Vacuum canisters used for trace gases collection are made from stainless steel
290 that is usually passivated following coating with a layer of chrome-nickel oxide (e.g. the
291 Summa coating technology) to reduce internal surface activity (Król et al., 2010a).
292 Pre-cleaned vacuum canisters can collect air samples passively via free flow under
293 atmospheric pressure or actively by using a pump (Fig. 4) (Ras et al., 2009). Samples
294 collected in these stainless steel canisters have a longer storage time than those taken by
295 sampling bags, and a decreased risk of chemical alteration. However, the high initial costs and
296 difficulties in cleaning and evacuating the canisters are usually the main reason for their
297 limited application.

298 **Glass-bombs.** Glass-bombs are specially designed glass bottles with air-tight valves to
299 control gas flow, similar to the canisters (Fig. 4). Before sampling, they are evacuated with a
300 vacuum pump, and air samples are collected passively via free flow under atmospheric
301 pressure. Glass-bombs have advantages over sampling bags and canisters in terms of sample

302 storage time and cost; however, trace gases might condensate onto the glass wall, and
303 glass-bombs are fragile and usually not compatible with modern analytical techniques. Hence,
304 their application in collecting trace gas samples from landfills is limited in the literature, and
305 they are more often used in odorous gas sampling for olfactometric analysis.

306

307 *****Table 2*****

308

309 *****Fig. 4.*****

310

311 *3.1.2. Sorbent trapping techniques*

312 Sorbent trapping is a well-established sampling technique which traps the trace
313 compounds onto various sorbent materials through adsorption or reaction with the sorbent
314 surface (Harper, 2000). Both active and passive sampling is possible when using sorbent
315 trapping techniques. For active sampling, an air pump is used to extract a defined volume of
316 air through a single or combined sorbent bed placed in a tube (sorbent tubes). For passive
317 sampling, trace compounds pass through a diffusion barrier via diffusion and permeation to
318 reach the collecting medium (diffusive tubes) (Fig. 4) (Król et al., 2010a). Time-integrated
319 samples can be obtained through sorbent trapping techniques, for either short- or long-term
320 sampling, which enables the calculation of average concentrations representing sampling time.
321 The storage time for air samples collected in sorbents is longer than for those collected by
322 whole-air sampling techniques – up to 30 days, if stored properly (Boeker et al., 2014; Brown
323 et al., 2014; Ho et al., 2018) – although it is still recommended to analyse the samples as soon
324 as possible after sampling (EPA TO 17). In addition, the convenience of the sorbent trapping
325 technique in sampling and analysis enables its combination with various equipment and
326 techniques for different purposes. For example, sorbent materials can be fitted to a gas probe
327 when taking samples from the landfill body or from beneath the landfill cover (Schuetz et al.,
328 2003; Young and Parker, 1983). More recently, sorbent tubes have been installed onto
329 unmanned aerial vehicles to determine VOCs emissions from open area sources (Zhou et al.,
330 2017), which could also be applicable to landfill trace gas measurements. Bag sampling,

331 followed by transferring into sorbent tubes for analysis, is also frequently reported in the
332 literature (Table S2). There are various types of sorbent-based sampling options; however, in
333 this section, the focus is on sorbent tubes and selected passive samplers (diffusive tubes),
334 which are the most widely used devices for collecting trace gas samples at landfills.

335 **Sorbent tubes.** The sorbent tubes used in active sampling can be classified into two
336 types, depending on the desorption method, namely solvent extraction tubes, where the
337 sorbent (often activated charcoal) is packed in a glass tube, and thermal desorption (TD) tubes,
338 where sorbents are packed in stainless steel or glass tubes (Woolfenden, 2010a). Both types
339 have been applied in trace gas sampling from landfills, although the latter is dominant due to
340 the operational convenience and enhanced sensitivity (Woolfenden, 2010a). The sampling
341 flow rate can vary from 10 – 200 mL min⁻¹ (typically set at 50 mL min⁻¹ for TD tubes), but
342 higher sampling rates can also be used for short-term monitoring. The sampling procedure has
343 been well-documented in different official methods, such as EPA TO-17, ASTM D-6196-97,
344 NOISH 2549, etc. (Ras et al., 2009; Woolfenden, 2010a). When a wide range of compounds
345 with different levels of volatility needs to be collected, multiple sorbent bed tubes can be used.
346 They are packed with two or more adsorbent materials with increasing adsorption strength in
347 a ‘sandwich’ structure, and the air passes through the tube from the end with the weaker
348 sorbent to the end with the stronger sorbent during sampling, so that less volatile compounds
349 are trapped on the weak sorbents, and very volatile compounds are trapped on the strong
350 sorbent. Based on the choice of sorbent, different groups of trace compounds can be collected
351 and concentrated. When selecting suitable sorbents for gas or air sampling, a range of factors
352 should be considered, such as sorbent strength, thermal stability, inertness and hydrophobicity,
353 artefact levels, etc. (Woolfenden, 2010b). Various commercial sorbent tubes are available
354 targeting different chemical compounds, thus reducing the time taken to select sorbents and
355 pack tubes.

356 **Diffusive (passive) samplers.** Diffusive (passive) samplers with particular sorbent
357 materials can be placed by a targeted emission source in a landfill (e.g. above the working
358 face) for a certain period for passive sampling (Reinhart et al., 1992). The trace gas molecules
359 reach the collecting medium after diffusion and permeation through a diffusion barrier,

360 namely a stagnated air layer or a non-porous membrane, according to Fick's first law of
361 diffusion (Cao and Hewitt, 1991). Various commercial diffusive samplers are available for
362 determining VOCs in air, such as Radiello® diffusive tubes, ORSA 5, Ultra, Waterloo
363 membrane samplers, etc. (Marć et al., 2015; Partyka et al., 2007; Ras et al., 2009). Generally,
364 diffusive samplers fall into two categories, depending on geometrical appearance: axial
365 diffusive samplers, which include tube-style and badge-style configurations with an axial
366 direction of diffusion and low uptake rates, and radial diffusive samplers, which often give a
367 much higher uptake rate, due to the larger surface area and shorter diffusion path (Fig. 4)
368 (Grosse and McKernan, 2014). Both types can be paired with a selected sorbent bed that is
369 compatible with solvent extraction or thermal desorption, and various samplers are
370 commercially available.

371

372 ***3.2. Analytical techniques***

373 Trace gas emissions from landfills are usually characterised by diverse composition and
374 low concentration levels in ambient air, and they can consist of hundreds of compounds with
375 concentrations at or below ppb level. Hence, reliable and precise analytical techniques are
376 required to identify and quantify as many compounds as possible within one set of analytical
377 instruments. The most widely used analytical techniques and equipment include gas
378 chromatography (GC) coupled with different detectors such as mass spectrometry (MS),
379 flame ionisation detection (FID), electron capture detection (ECD) and pulsed flame
380 photometric detection (PFPD) (Ras et al., 2009). Ex-situ equipment placed in a laboratory is
381 the most preferable choice, and it requires additional sampling and/or pre-concentration
382 procedures and transportation of air samples from the field to the laboratory for analysis. The
383 optimal analytical method is characterised by low detection limits (generally below ppb or
384 even ppt levels) with good separation efficiency of the individual gas species. However, it can
385 be complicated and time-demanding to develop the analytical method on different types of
386 equipment, for example a TD-GC-MS with a selection of suitable GC columns, the
387 determination of working conditions for TD, GC and MS, calibration and quality control and,
388 if necessary, system performance improvement.

389 Continuous monitoring with ex-situ techniques is difficult, due to the large number of
390 samples required, as well as high cost. In addition, the storage and transportation of samples
391 can also generate problems. On-site equipment, on the other hand, combines the sampling and
392 analysis processes into one set of portable instruments, thereby permitting on-line monitoring
393 in the field. Several applications of miniature chromatographic systems (μ GC-MS coupled
394 with thermal desorption pre-treatment) have been used for measuring trace gas samples at
395 landfills (Davoli et al., 2003; Liu et al., 2015; Sadowska-Rociek et al., 2009). Publications on
396 other portable instruments enabling real-time analysis, such as selected ion flow tube mass
397 spectrometry (SIEF-MS) and online TD - GC - time-of-flight mass spectrometry (TOFMS),
398 are also available (Boeker et al., 2014; Langford et al., 2014; Ras et al., 2009). Reviews of
399 different on-line gas analysers for monitoring VOCs in air have been published elsewhere
400 (Król et al., 2010b; Marć et al., 2015). Compared to ex-situ techniques, on-site practices tend
401 to have higher detection limits (e.g. at ppb or ppm levels) and larger variability (Harshman et
402 al., 2017). Additionally, fewer compounds can be detected and quantified, and their separation
403 is also often poor, due to the short analytical time (Marć et al., 2015). These limitations have
404 restricted the use of portable instruments for measuring trace gas emissions from landfills,
405 and further developments are still required, in order to meet user requirements for analysis
406 precision and convenience.

407 It should be noted that the choice of sampling and analytical method will affect the
408 conclusions in a study. As the number of detectable compounds is often limited due to the
409 restriction of instrument selectivity and sensitivity, it is easy to conclude some compounds are
410 dominant in a particular landfill, whereas the real situation might differ. For example,
411 ammonia and hydrogen sulphide are usually present in LFG at considerable concentrations;
412 however, they are not always included in studies concerning trace gas emission from MSW
413 landfills since their analysis requires additional analytical instruments, whereas most VOCs
414 can be measured simultaneously by GC-MS. In addition, when sorbent tubes are used in
415 sample collection, some very volatile compounds such as chlorofluorocarbons (CFCs) might
416 not be well trapped onto the sorbent, which could also lead to a bias in understanding the
417 trace gas composition at a landfill site. Hence, whenever a research study is conducted with

418 the purpose of investigating trace gas emission from landfills, the restrictions of the sampling
419 and analytical techniques applied and their potential effects on the reported data have to be
420 clearly reflected in the conclusion. This also applies to the data compiled in Section 5 and
421 Section 6, and an example will be given in Section 6.

422

423 **4. Quantitative measurement of trace gas emissions from MSW landfills**

424

425 Emission rates or surface emission fluxes are often necessary to determine environmental
426 impacts and human health risks, as well as to monitor the results of pollution control activities
427 (Canzano et al., 2010; Kim, 2016). Emission rates are also input data for most air dispersion
428 models (including odour dispersion models) when estimating the downwind concentration of
429 certain pollutants and defining the safe distance between landfills and residential areas (De
430 Melo Lisboa et al., 2006; Murthy and Mcfarland, 1993). In addition, knowing the emission
431 rates of trace gases could support decision-making procedures for the construction of new
432 waste management facilities, since they are important input data for life cycle assessment
433 (LCA) models (Alfonsín et al., 2015).

434 Many researchers have conducted laboratory- and pilot-scale studies in reactors to
435 simulate trace gas emissions from MSWs under various conditions (Canzano et al., 2010;
436 Pierucci et al., 2005; Staley et al., 2006; Tan et al., 2017). However, considering the
437 complexity of real landfill conditions, these data cannot be used in estimating or predicting
438 emissions. Very limited data can be found regarding the quantitative measurement of trace
439 gas emission rates from MSW landfills (see later in Section 6). In fact, the quantitative
440 measurement has proven to be extremely challenging, due to huge temporal and spatial
441 variations, as well as the technical limitations in sampling and analysis. Several techniques
442 have been developed in this regard, albeit they all have limitations, and none of them has been
443 accepted universally as the best way to perform such measurements.

444 Existing quantitative measurement techniques can be categorised as either direct or
445 indirect (Hudson and Ayoko, 2008a; Reinhart et al., 1992). Direct measurement usually
446 involves taking field samples from gas vents or above landfill surfaces, followed by chemical

447 analysis that enables emission rate calculation. Indirect measurements mainly provide
448 emission rates via modelling, using input data such as measured or calculated trace gas
449 concentrations in ambient air above or downwind of the landfill. Direct techniques, in
450 particular surface flux chambers, are widely used in measuring fugitive emissions from
451 specific landfill surfaces, which normally give ‘real’ emission data, whereas most indirect
452 techniques are used for estimating whole-site emissions. A detailed introduction to those
453 techniques is given below.

454

455 **4.1. Direct measurement techniques**

456 *4.1.1. Gas vent sampling and velocity measurement*

457 For landfills with LFG collection systems, the total emission rate of trace compounds can
458 be estimated by measuring the trace gas concentrations in raw LFG and the volumetric flow
459 rate of LFG from the vent pipe. Gas samples for concentration measurements are taken at the
460 ends of active or passive vent pipes (before flaring or other treatments) with measurable gas
461 flows. The sampling is often conducted by bag sampling. The emission rate is calculated as
462 follows:

$$463 \quad E_i = Q_{LFG} \cdot C_i \quad (\text{Equation 1})$$

464 where E_i is the emission rate of the trace compound i ($\mu\text{g s}^{-1}$), Q_{LFG} is the volumetric flow
465 rate of LFG in the gas vent ($\text{m}^3 \text{s}^{-1}$) and C_i is the concentration of trace compound i in LFG
466 ($\mu\text{g m}^{-3}$). Sometimes, unit conversion is needed if the pressure and temperature in the vent
467 pipe differ significantly from the ambient air.

468 Saral et al. (2009) used this method to calculate the emission rates of selected VOCs
469 from a landfill and then employed them as input data for an atmospheric dispersion model
470 (Industrial Source Complex v3 Short Term model, ISCST3). Instead of directly measuring
471 trace gas concentrations by taking air samples at the vent pipe, they used the default
472 concentration data (emission factor) for trace gases in LFG provided by the US EPA (1995).
473 The volumetric VOC flow rate from each gas vent was calculated by multiplying the
474 corresponding emission factors and the gas flow rate of each vent, and these were then
475 transferred into mass flow rates, using the ideal gas law. The overall emission rate for the

476 whole landfill area was assumed as the sum of the calculated emission rate from all gas vents.
477 Similar applications can also be found in other studies estimating emission rates based on
478 measured trace gas concentrations and LFG exit velocity from vents pipes (Allen et al., 1997;
479 Kim et al., 2006). In addition, emissions from flares and gas engines can also be calculated in
480 this way by simply measuring trace gas concentrations in the exhaust air stream and the
481 emission air flow rate from flares and engines during standard working procedures (Palmiotto
482 et al., 2014).

483 This method is simple and can give both concentration (if measured) and emission rates.
484 However, the calculation assumes that all of the extracted LFG is emitted directly into the
485 atmosphere. Hence, when there is an LFG flaring or utilisation system, in which most trace
486 gases are destroyed or removed, applying this method would lead to significant bias. In
487 addition, fugitive emissions from landfill surfaces, and the trace gases contained in
488 non-collected LFG, are not included in this method, which might comprise a significant part
489 of the total trace gas emissions and thus further increase the uncertainty attached to this
490 method.

491

492 *4.1.2. Surface flux chambers*

493 Surface flux chambers are enclosure devices that are placed on a defined surface area.
494 The trace gases emitted from the surface of a landfill are captured inside the chamber, and by
495 monitoring the concentration changes and the airflow through the device (if used), the surface
496 emission rates can be calculated (Mønster et al., 2019). There are different variations of
497 surface flux chambers, but in general they can be classified into static chambers, dynamic flux
498 chambers and wind tunnels, depending on the way they are operated and the physical
499 configuration of the device (Gostelow et al., 2003; Hudson and Ayoko, 2008b; Mønster et al.,
500 2019). Fig. 5 shows the concepts of a static chamber, a dynamic flux chamber and a wind
501 tunnel, including the basic equations used for calculating emission rates in each case (Mønster
502 et al., 2019). Detailed introduction to those techniques can be found in the above cited papers.

503

504 *****Fig. 5.*****

505

506 The static chamber is by far the most frequently used technique for measuring trace gas
507 emission rates from landfill surfaces (e.g. Bogner et al., 1997; Schuetz et al., 2003, 2008;
508 Tassi et al., 2010; Yesiller et al., 2018; Wang et al., 2019), and there is very little emission
509 dilution when one is used, which is good for measuring surfaces with low emission rates.
510 However, pressure increases might happen inside the chamber during its installation, and the
511 accumulation of trace compounds concentrations in the headspace may suppress gas
512 transportation from the landfill surface to the chamber (Hudson and Ayoko, 2008b), resulting
513 in an underestimation of the actual emission rate. In addition, when whole-air sampling
514 techniques (bags or canisters) are used for taking samples from the chamber, a large sample
515 volume is often required, in order to reach the instrumental detection limits due to low
516 concentrations of trace gases, and this can lead to significant pressure fluctuations in the
517 chamber and influence the emission rate.

518 To avoid pressure build-up inside the static chamber, different methods have been
519 developed, such as collecting the samples over a short time period after installation (e.g. < 30
520 min) (Tassi et al., 2010), adding a long small-diameter vent tube to the chamber (Mønster et al.,
521 2019), using parallel or large-scale chambers (Bogner et al., 1997; Mønster et al., 2019;
522 Scheutz et al., 2008; Schuetz et al., 2003; Yesiller et al., 2018), or adding a small
523 compensation air flow of clean inert gas into the chamber at the sampling rate (de Sá Borba et
524 al., 2017; Gallego et al., 2014; Gostelow et al., 2003; Martí et al., 2014). However, those
525 methods might have a risk of introducing other errors, and underestimation of the actual
526 emission rate might still happen due to the concentration accumulation of trace gases inside
527 the chamber and the decrease of the concentration gradient between the landfill surface and
528 the chamber headspace (Gostelow et al., 2003).

529 The dynamic flux chamber is operated under a fixed, controlled air flow, thus reducing
530 the risk of pressure build-up inside the chamber. This method is recommended in the USA for
531 measuring gaseous emission rates from solid surfaces (Klebusch, 1986), and guidance on the
532 design and operation of dynamic flux chamber has been provided by some researchers for
533 measuring fugitive VOC emissions from landfill surfaces (Eklund, 1992; Reinhart et al.,

534 1992). However, only a few applications were reported in previous studies that used dynamic
535 flux chamber to measure VOC emissions from landfill sites (González et al., 2013; US EPA,
536 1995).

537 One challenge of using dynamic flux chamber is that it is difficult to identify when the
538 concentration inside the chamber has reached a steady state, due to the lack of real-time
539 measuring instruments for trace gases (often, discrete air samples are extracted and analysed
540 later in the laboratory). In addition, the measured emission rate is dependent on the used air
541 flushing rate, and any underestimation or overestimation of the actual emission rate might
542 occur as a result of improper flushing flow rates (Eklund, 1992; Kolari et al., 2012; Zhang et
543 al., 2002). Hence, validation is required for obtaining the optimal operation conditions for a
544 dynamic flux chamber. Capelli et al. (2014a) tested a dynamic flux chamber and a static
545 chamber for measuring methane emission rates from landfill surfaces, and they found good
546 agreement between results measured by the two devices. However, no such comparison was
547 made in literature between the two types of chambers for measuring trace gas emissions, and
548 further investigations are needed to standardise the emission rate measurements by dynamic
549 flux chambers.

550 Wind tunnels are similar to dynamic flux chambers in principal, albeit wind tunnels use
551 much higher air flow rates (typically with a sweeping velocity of $0.1 - 5 \text{ m s}^{-1}$) and have a
552 prismatic measurement section installed with the inert airflow sweeping longitudinally along
553 the tunnel (Gostelow et al., 2003), which allows for simulating surface emissions under
554 different wind actions. There are many different configurations of wind tunnels (Gostelow et
555 al., 2003), and Fig. 5 shows an example of the UNSW tunnel developed by Jiang et al. (1995),
556 which has been applied in measuring landfill surface emissions. The calculation of the
557 emission rate derived from the wind tunnel is given below (Equation 2) (Hudson and Ayoko,
558 2009):

559
$$E_i = C_i \cdot v_1 \cdot \frac{A_1}{A_2} \quad (\text{Equation 2})$$

560 where E_i is the emission rate of trace compound i ($\mu\text{g m}^{-2} \text{ min}^{-1}$), C_i is the measured
561 concentration of trace compound i ($\mu\text{g m}^{-3}$) at a steady state, v_1 is the sweeping air flow rate
562 inside the wind tunnel (m s^{-1}), which is normally estimated by Q/A_1 , Q is the flushing flow

563 rate ($\text{m}^3 \text{s}^{-1}$), A_1 is the cross-sectional area of the wind tunnel (m^2) and A_2 is the surface
564 area covered by the wind tunnel (m^2). If the obtained emission rates are to be used as inputs
565 for dispersion modelling, they need to be recalculated at the correct wind speed for each hour
566 of the simulation, which can be done by using several different methods (F. Lucernoni et al.,
567 2017). The most commonly applied approach for this re-calculation is provided as Equation
568 (3) (F. Lucernoni et al., 2017):

$$569 \quad E_{v_2} = E_{v_1} \left(\frac{v_2}{v_1} \right)^{\frac{1}{2}} \quad (\text{Equation 3})$$

570 where E_{v_2} is the recalculated emission rate of trace compound i ($\mu\text{g m}^{-2} \text{min}^{-1}$), E_{v_1} is the
571 measured emission rate by wind tunnel ($\mu\text{g m}^{-2} \text{min}^{-1}$) at a sweeping air flow rate v_1 (m s^{-1})
572 and v_2 is wind speed at a specific hour of the simulation domain, which is conventionally
573 measured at 10 m above ground (m s^{-1}).

574 The wind tunnel has been used widely in measuring odours and VOC emission rates
575 from liquid and solid area surfaces such as lagoons and animal feed yards (e.g. Sironi et al.,
576 2005; Jiang and Kaye, 1996; Capelli et al., 2008; Hudson and Ayoko, 2008b; Capelli et al.,
577 2013a; D. B. Parker et al., 2013; Palmiotto et al., 2014; F. Lucernoni et al., 2017). However,
578 only one application was found in literature, in which a wind tunnel was used for measuring
579 fugitive trace gas emission rates from the working face of two landfills in China (Liu et al.,
580 2015; 2016b). A more detailed discussion on the data will be offered in Section 6.

581 Similar to the dynamic flux chamber, the emission rates generated from a wind tunnel are
582 highly dependent on the adopted wind sweeping speed as well as the physical dimension of
583 the tunnel (Hudson and Ayoko, 2008a). Understanding the aerodynamic performance of the
584 tunnel is very important when carrying out measurements (Scotto di Perta et al., 2016), and
585 uniform concentration and air velocity profiles have to be developed therein, to avoid possible
586 errors and to obtain representative measurements that can simulate the real field conditions at
587 the emission surface (Gostelow et al., 2003). However, it is often difficult to verify
588 aerodynamics inside a wind tunnel, thereby leading to unknown uncertainties in the measured
589 emission rates. Those measured by wind tunnels are often very different from those measured
590 by other devices in previous studies, as summarised by Hudson and Ayoko (2008b). In fact,
591 recently, the application of wind tunnels on landfill surfaces has been questioned, since

592 significant overestimations of the real emission rates have been uncovered (Capelli and Sironi,
593 2018; F. Lucernoni et al., 2017). The application of wind tunnels assumes a passive surface
594 and the emission as a function of wind speed (F. Lucernoni et al., 2017). However, this is not
595 true for landfill surfaces, since the emission of trace gases from landfill surfaces is mainly
596 governed by the LFG emission rate and is affected by many other factors as has been
597 discussed in Section 2.2. It may therefore be necessary to introduce some conversion factors
598 into the emission rate calculation when applying wind tunnels on solid surfaces (Capelli et al.,
599 2012; D. Parker et al., 2013), and a validation test is needed in real field conditions. In
600 addition, the uncertainty range of this method needs to be established.

601 Despite the limitations of the three measurement devices mentioned above, the
602 application of surface flux chambers at landfills is often restricted, due to the large temporal
603 and spatial emission variations as well as the indefinite geometry and heterogeneous surfaces
604 (Mønster et al., 2019; Murthy and Mcfarland, 1993). Many measurements are required to
605 obtain a reliable estimation of the total trace gas emission from a landfill surface; however,
606 this can be very time- and labour-consuming, and is often impossible in practice. In addition,
607 surface flux chambers cannot be used to measure emissions from cracks in surface covers and
608 leaks in gas and leachate collection systems. A more detailed description of the advantages
609 and limitations of surface flux chambers has been provided by Mønster et al. (2019).

610

611 **4.2. Indirect measurement techniques**

612

613 *4.2.1. LFG production modelling*

614 This method calculates the emission rate by multiplying the LFG production rate with
615 trace gas concentrations in raw LFG. The LFG production rate is estimated by applying
616 various LFG production models, such as LandGEM, the IPCC model or others. The trace gas
617 emission rate is given as follows (Paraskaki and Lazaridis, 2005):

$$618 \quad E_i = \frac{Q \cdot C_i}{10^9} \quad (\text{Equation 4})$$

619 where E_i is the emission rate of trace compound i (kg year^{-1}), Q is the LFG generation rate
620 ($\text{m}^3 \text{ year}^{-1}$) calculated by an LFG production model (e.g. LandGEM) and C_i is the

621 concentration of trace compound i in raw LFG ($\mu\text{g m}^{-3}$) derived from the literature or from
622 field measurements.

623 This method is simple and has been widely used in providing trace gas emission rates for
624 dispersion models with the purpose of assessing environmental impacts and health risks (e.g.
625 Tagaris et al., 2003; Paraskaki and Lazaridis, 2005; Pagé et al., 2007; Moolla et al., 2013; Cai
626 et al., 2015; Abdul-Wahab et al., 2016; Araiza-Aguilar and Rojas-Valencia, 2018). In the cited
627 papers, emission rates of target odorous or toxic trace compounds were calculated by using
628 various LFG production models and applying Equation (4). Most of the studies applied
629 default LFG trace gas concentrations rather than site-specific trace gas concentrations based
630 on measurements. The obtained emission rates were put into an air dispersion model to
631 determine the odour/health risk critical zone around the landfill. This method is also
632 applicable for decision-making processes.

633 It should be noted that the LFG production models can be highly uncertain and are
634 designed to estimate raw LFG generation instead of emission. Hence, in landfills where part
635 of the trace gases are collected and destroyed through an LFG collection system, this method
636 might significantly overestimate overall trace gas emissions, unless the model is adjusted for
637 collected LFG. When trace gas concentration data are obtained from the literature, for
638 example if default NMOC concentrations under AP-42 (USEPA) are used, trace gas emissions
639 are most likely overestimated, as these data are not representative of current landfill practices
640 – at least in many Western countries where, for example, hazardous waste is not disposed in
641 landfills (Hamideh Soltani-Ahmadi, 2000; Staley et al., 2006). In addition, it is not reasonable
642 to use a constant concentration of trace gases over the entire decomposition cycle (Staley et
643 al., 2006), whilst the adsorption, transformation and degradation of trace gases are not
644 considered in this method, as they increase uncertainty even further.

645

646 4.2.2. *Estimation by using relative concentration ratios to CH₄*

647 Another indirect measurement method is to first measure the fugitive CH₄ emission rate
648 with different techniques, for example a surface flux chamber, and then calculate the trace gas
649 emission rate by using relative concentration ratios to CH₄ in raw LFG or in ambient air

650 measured above landfill surfaces, as shown in Equation (5):

651
$$E_i = \frac{E_m \cdot C_i \cdot 22.4}{C_m \cdot M_m} \cdot 10^{-3} \quad (\text{Equation 5})$$

652 where E_i is the emission rate of trace compound i ($\mu\text{g m}^{-2} \text{d}^{-1}$), E_m is the measured emission
653 rate of CH_4 ($\text{g m}^{-2} \text{d}^{-1}$), C_i and C_m are the concentrations of trace compound i ($\mu\text{g m}^{-3}$) and
654 CH_4 (expressed as volume percentage) measured in raw LFG or in ambient air measured
655 above landfill surfaces, M_m is the molecular weight of CH_4 (16.04 g mol^{-1}), 22.4 is the
656 standard volume of an ideal gas at 1 atmosphere pressure (L mol^{-1}) and 10^{-3} is a conversion
657 factor.

658 Several applications of this method were found in the literature. For instance, in a project
659 conducted by a UK company which aimed at managing odour risks at landfill sites
660 (McKendry et al., 2002), the odorous gas emission rate was calculated by using published
661 data with the ratio of odorant to CH_4 in raw LFG, while CH_4 emission rates were measured by
662 static chambers. Yue et al. (2014) also used a static chamber to measure CH_4 emission rates
663 from the operating area, inoperative area and soil cover of a landfill in China. The
664 concentration ratios between trace gases and CH_4 were obtained by taking ambient air
665 samples above the monitored surfaces, and the emission rates of sulphur compounds were
666 calculated by Equation 5 herein. In another study conducted in the US, the Vertical Radial
667 Plume Mapping method was applied to measure fugitive CH_4 emissions from three landfills
668 (G&M, 2012). Raw LFG samples were collected from the gas header pipe at each site in
669 order to obtain concentration ratios of trace compounds and CH_4 , and the fugitive emission
670 rates of target trace compounds from monitored landfill areas were calculated.

671 In theory, all of the methods presented by Mønster et al. (2019) for measuring fugitive
672 CH_4 emissions from landfills can form the basis for using Equation 5. When downwind plume
673 measuring techniques are used in CH_4 emission measurement, such as the tracer dispersion
674 method and the differential absorption LiDAR method, whole-site fugitive emission rates can
675 be acquired, which is advantageous compared to surface flux chambers. However, it remains
676 a challenge to establish a constant concentration ratio between trace gases and CH_4 , due to
677 temporal and spatial variations in trace gases and CH_4 concentrations. Trace gas-to- CH_4 ratios
678 are expected to vary depending on waste composition and age. Even when site-specific

679 samples are collected to obtain concentration ratios, there are still unknown uncertainties in
680 the results estimated by this method. In particular, care has to be taken when using
681 concentration ratios in raw LFG for estimating fugitive trace gas emission rates, since trace
682 gases and CH₄ undergo different transformations and oxidation during transportation, causing
683 different ratios in ambient and raw LFG samples.

684

685 *4.2.3. Inverse dispersion modelling*

686 The inverse dispersion modelling method estimates trace gas emission rates via air
687 dispersion models by using ambient concentrations of trace gases measured downwind of the
688 landfill and meteorological parameters as inputs (Lehning et al., 1994; Rao, 2007). Various
689 models can be used in this method, including some meteorological models and air dispersion
690 models, most of which are Gaussian models.

691 The inverse dispersion modelling method enables the measurement of overall emission
692 rates across a large surface area and has been frequently applied for measuring CH₄ and odour
693 emissions from landfill sites (e.g. Nicholas Scott Guarriello, 2007; Sarkar and Hobbs, 2003;
694 Soriano et al., 2007). However, there are only a few applications in estimating trace gas
695 emission rates. In the study conducted by Schauburger et al. (2011), ambient concentrations of
696 several selected odorous compounds were measured downwind of a thermal waste recycling
697 plant, together with meteorological parameters (wind velocity, wind direction and the stability
698 class). By applying the inverse dispersion modelling method, using the Austrian regulatory
699 Gaussian model, they calculated the overall odour emission rate at the source by adding up
700 the back-calculated emission rate of the selected odorants. Similarly, Davoli et al. (2010) also
701 used the inverse dispersion modelling method to estimate the surface emission rate of vinyl
702 chloride monomer from a landfill in a study aiming at health risk assessment.

703 The inverse dispersion modelling method is useful when in situ measurement via direct
704 techniques at the emission source is not available. It can also be used to validate the results
705 obtained from direct measurements. However, this method requires accurate field
706 measurement of ambient concentrations of trace gases (Rao, 2007), which is often constrained
707 by the limited number of samples and the location of sampling points in the downwind

708 direction. In addition, the accuracy of this method is significantly affected by meteorological
709 and site-specific factors as well as the quality and appropriateness of the model used (Laor et
710 al., 2014), which makes it difficult to establish an uncertainty range for this method.

711

712 **5. Trace gas constituents and concentrations in MSW landfills**

713 According to the relevant information given in the literature (2003-2019), concentration
714 data of 63 common trace compounds are compiled in this section (see Table S1). Previous
715 studies have reported concentration data for hundreds of trace compounds; however, in this
716 paper it was decided to focus on those 63 compounds as they were frequently reported in
717 literature and their concentrations contribute most to the total trace gas concentrations. Hence,
718 the conclusions made in this section are still considered representative for MSW landfill sites
719 on a generic basis, though bias might exist for site-specific cases.

720 The concentration data are classified into two major groups: 1) concentrations measured
721 in raw LFG samples obtained from LFG collection systems (active/passive) or at LFG
722 leakage points and 2) concentrations in ambient air at different landfill units, measured above
723 surfaces/areas, defined as active working areas and uncovered surfaces (UN), temporally
724 covered surfaces (TC), final covers (FC), leachate-related areas (LC, leachate
725 collection/storage pool or treatment plant) and the overall site (WH, whole-site average
726 concentrations). Detailed concentration data and landfill information can be found in the
727 Supplementary Material (SM) (Table S1 and S2).

728 ***5.1. Trace gas concentrations in raw LFG and in ambient air at different landfill units***

729 Fig. 6 shows the reported concentrations of different chemical groups in raw LFG and in
730 ambient air at the above-defined landfill units. Total trace gas concentrations in raw LFG
731 normally range from 10^3 to 10^7 $\mu\text{g m}^{-3}$ (Fig. S1), whereas concentrations in ambient air are
732 about 3 – 4 orders of magnitude lower., and sulphur compounds and aromatics are the most
733 abundant chemical groups, followed by aliphatic hydrocarbons (average concentrations
734 between 10^5 – 10^6 $\mu\text{g m}^{-3}$). Oxygenated compounds, terpenes and halogenated compounds
735 have been measured in lower concentrations (about one order of magnitude lower than the
736 other three groups). High concentrations of nitrogen compounds (mainly ammonia) have also

737 been detected occasionally; however, the limited amounts of data do not support a solid
738 conclusion in this regard. It should be mentioned that the concentrations compiled in Fig.
739 6 were measured in different landfills around the world with very diverse waste composition,
740 climatic condition and landfill management operation, and different sampling/analytical
741 techniques were used (see Table S2). In addition, this manuscript only considered 63
742 different compounds commonly measured in landfill studies. Hence, the relative abundance of
743 each chemical group and the concentration range exhibited in Fig. 6 represent a general trend
744 but do not necessarily apply to site-specific situations.

745

746 *****Fig. 6.*****

747

748 High ambient trace gas concentrations are mostly found in active working areas and
749 uncovered waste surfaces (UN), which are normally associated with fresh waste in the initial
750 adjustment and transition stages. Oxygenated compounds make up the dominant chemical
751 group, and these are typical products generated from the degradation of fresh waste. In
752 addition, high concentrations of aromatics are often detected, most likely originating from
753 volatilisation of chemicals in waste products as well as from diesel combustion in the waste
754 trucks and operation machines (if the measurement is taken on the active dumping area) (Yao
755 et al., 2015). In addition, the older waste dumped beneath the fresh waste could also
756 contribute to the trace gas emission from the uncovered surface. Hence, the elevated trace gas
757 concentrations in uncovered surfaces are caused by a combination of emissions from fresh
758 waste, underlying old waste and waste operation vehicles. The total concentration of trace
759 gases on covered surfaces is lower compared to uncovered surfaces (Fig. S1), and the
760 concentration of oxygenated compounds is significantly lower. However, relatively high
761 concentrations of sulphur compounds, aromatics and terpenes are still measured.
762 Leachate-related areas are dominated by oxygenated compounds and ammonia, which are
763 typical compounds emitted from this source.

764 In the following sections, we will discuss in detail the origin and concentration
765 distribution of common trace compounds in LFG and in ambient air for each chemical group,

766 as defined in Table 1.

767

768 **5.2. Sulphur compounds**

769 Sulphur compounds are generated either by microbial sulphate reduction, from
770 sulphate-containing waste products such as gypsum drywall, or via the microbial
771 decomposition of organic sulphur-containing waste types, for example the anaerobic
772 degradation of food waste (Chen et al., 2017; Ko et al., 2015; Zhang et al., 2013). Some
773 organic sulphur compounds such as dimethyl sulphide (DMS) and dimethyl disulphide
774 (DMDS) might also originate from the direct volatilisation of inherited residuals such as
775 aromatic compounds in food products, but this amount is insignificant compared to generation
776 as a result of microbial sulphate reduction or waste decomposition processes (Wu et al., 2010).
777 Large amounts of organic sulphur compounds are produced and emitted during the early
778 stages of waste decomposition (Fig. 3) (Kim, 2006; Lou et al., 2015; Zhang et al., 2013), and
779 they are thought to be indicative of relatively fresh waste (Duan et al., 2014; Young and
780 Parker, 1983; Zhao et al., 2015). During the methanogenesis stage, more H₂S is produced and
781 emitted (Fig. 3), and in conditions with high temperatures or at landfills with leachate
782 recirculation, higher production and emissions of sulphur compounds are usually observed,
783 due to enhanced microbial activities (Kim, 2006; Ko et al., 2015; Moreno et al., 2014; Tan et
784 al., 2017).

785 Sulphur compounds are one of the dominant chemical group in raw LFG, and H₂S
786 usually prevails over other sulphur compounds (Fig. S2). Kim et al. (2005, 2006) measured
787 concentrations of reduced sulphur compounds (H₂S, methyl mercaptan, DMS, DMDS and
788 carbon disulphide) from vent pipes at six Korean landfills and found that H₂S accounted for
789 more than 90% of the sulphur-containing gases in LFG, whilst average concentrations ranged
790 across several orders of magnitude, from 6.3 µg m⁻³ up to 3.8·10⁶ µg m⁻³. The low
791 concentrations (below 10 µg m⁻³) in this regard were measured at closed or old landfills (Kim,
792 2006), whereas young and active landfills were responsible for the higher concentrations
793 ranging from hundreds of thousands to several millions. Comparable concentrations were also
794 reported by Moreno et al. (2014), who measured H₂S concentrations as 2.7·10⁶ µg m⁻³ in raw

795 LFG from gas leakage points (e.g. cracks on soil cover or splits in the geomembrane). When
796 large amounts of construction and demolition (C&D) waste are disposed at a landfill, H₂S
797 production can be significantly enhanced, due to the high sulphate content in gypsum drywall
798 (or wall board) (Lee et al., 2006), which in an extreme case could result in up to 35% of LFG
799 volume in an old UK landfill, as reviewed by Parker et al. (2002). Methyl mercaptan and
800 DMS are the dominant organic sulphur compounds in raw LFG, normally with concentrations
801 below $5 \cdot 10^4 \mu\text{g m}^{-3}$ in active landfills (Fig. S2). It is worth mentioning that Fang et al. (2012)
802 and Cheng et al. (2018) reported much lower concentrations in raw LFG compared to other
803 studies (also for other trace compounds), since their samples were collected from open
804 passive vents installed in cells with aged waste that had almost reached the stabilisation stage
805 (personal communication with the authors). Hence, both air dilution and the age of the waste
806 contributed to the low concentrations they reported.

807 Concentrations of sulphur compounds in ambient air measured above different landfill
808 surfaces/areas are generally below $1000 \mu\text{g m}^{-3}$, and the highest concentrations are mostly
809 observed at the active working area or above uncovered waste surfaces (Fig. S3). Of those
810 gases measured above landfill surfaces, H₂S also appears frequently in high concentrations
811 (up to $800 \mu\text{g m}^{-3}$), although abundant levels of carbon disulphide, DMS and DMDS are also
812 found, sometimes even exceeding H₂S concentrations and becoming the dominant compounds
813 (Table S1) (Duan et al., 2014; Shon et al., 2005; Song et al., 2007; Yue et al., 2014).

814

815 ***5.3. Oxygenated compounds***

816 Oxygenated compounds are mostly of biogenic origin and consist of a wide variety of
817 compounds. Alcohols and ketones can be produced as aerobic metabolism intermediates as
818 well as under anaerobic conditions. Incomplete aerobic degradation can also lead to the
819 production of alcohols, aldehydes, esters, ethers and acids. During waste collection,
820 transfer/transportation and disposal processes, aerobic and incomplete-aerobic conditions
821 develop and large amounts of the above-mentioned compounds are produced (mainly alcohols)
822 (Fig. 3) (Statheropoulos et al., 2005; Tan et al., 2017). As the waste enters the acidogenesis
823 and acetogenesis stages, volatile fatty acids (VFAs), especially acetic acid, butyric acid and

824 ketones, become dominant, due to the hydrolysis of organic matter in the landfilled waste.
825 Concentrations of oxygenated compounds in raw LFG drop sharply after the methanogenesis
826 stage begins (Fig. 3). Solvents and plastic packaging can also be sources of alcohols and
827 ketones in landfills (Staley et al., 2006), although their contributions might be small compared
828 to the biological degradation of organic materials.

829 Concentration data for oxygenated compounds are limited in published studies, and they
830 are often not comparable with each other, since they focus on different compounds. In Fig. S2,
831 concentrations of several representative compounds in this group are summarised, and it
832 seems that the most abundant oxygenated compounds in raw LFG are ketones, ethyl acetate
833 and VFAs, with average concentrations between $1.1 \cdot 10^3$ and $3.8 \cdot 10^4 \mu\text{g m}^{-3}$, whilst acetone is
834 the dominant compound. VFA, ketones and alcohols are typical products produced by the
835 hydrolysis of the organic waste fraction (acidogenesis stage), and acetate is produced by
836 acetogenic organisms originating from organic acids (Angelidaki and Batstone, 2011). In
837 addition, an accelerated concentration of ketones might be observed when abundant plastic
838 waste is landfilled, since ketones are often present in plastic packaging (Hodgson et al., 2000).
839 VFAs are water-soluble and might dissolve into leachate instead of being released into the gas
840 phase, which could explain the higher concentration of acetic acid in leachate-related areas
841 (Fig. S4).

842 Alcohols, primarily ethanol, which are typical products resulting from incomplete
843 aerobic degradation processes, turn out to be the dominant compounds in ambient air in most
844 cases, especially in the active working area (Fig. S4) (Duan et al., 2014; Wu and Wang, 2015),
845 and they are likely to be indicative of relatively fresh waste that is in the early stages of
846 decomposition (Young and Parker, 1983). The concentration of ethanol can be as high as
847 $2.4 \cdot 10^3 \mu\text{g m}^{-3}$ at the working face (Table S1 in SM). Also, relatively abundant levels of
848 acetone, ethyl acetate and acetic acid have been measured in the active working area,
849 normally at concentrations below $100 \mu\text{g m}^{-3}$, though high concentrations up to $6 \cdot 10^3 \mu\text{g m}^{-3}$
850 for acetic acid have also been reported (Fang et al., 2012).

851

852 **5.4. Aromatics**

853 Most aromatics are xenobiotic and originate through direct volatilisation from the
854 disposed waste. Such compounds can be released from paper waste, plastic packaging,
855 solvent-containing waste, petroleum products, etc., but they can also be emitted from food
856 waste, since some of them are responsible for food flavours or used as food additives
857 (Komilis et al., 2004; Mustafa et al., 2017). In addition, they can be formed during the
858 anaerobic degradation of lignin (Wilber and Murray, 1990), and it has been reported that high
859 fat content foods tend to release higher concentrations (Heikes et al., 1995), probably due to
860 the fact that most aromatics are lipophilic compounds and can be absorbed by lipids (Ham
861 and Komilis., 2003). Among the various origins, direct volatilisation from chemicals in the
862 disposed waste is considered the main source of aromatic compounds emitted from landfills
863 (Powell et al., 2006). Aromatics are an important chemical group in landfill environment –
864 often, they are the second largest group of trace gases present in LFG after sulphur
865 compounds, and sometimes they even dominate when H₂S concentration is low (Fig. 6)
866 (Gallego et al., 2012; Zou et al., 2003). Abundant concentration data on aromatics in raw LFG
867 have been reported in previous studies (Table S1).

868 Benzene, toluene, ethylbenzene and m,p,o-xylene (BTEX) dominate this group of trace
869 gases, in both raw LFG and in ambient air above landfill surfaces, and toluene is typically the
870 most abundant compound among them (Fig. S2 and Fig. S5). In young active landfills, the
871 highest concentration of toluene in raw LFG has been measured at nearly $3 \cdot 10^6 \mu\text{g m}^{-3}$ (Tassi
872 et al., 2009), while the other aromatics have been found in a range of 10^3 to $10^5 \mu\text{g m}^{-3}$ (Table
873 S1 in SM). Their concentrations will decrease along with landfill aging, reaching probably
874 below $100 \mu\text{g m}^{-3}$ at very old sites (Kim et al., 2006; Kim, 2006). This characteristic might be
875 useful for determining the landfill stabilisation stage.

876 Concentrations of individual aromatic compounds in ambient air measured above
877 different landfill units are generally below $100 \mu\text{g m}^{-3}$, still with toluene as the most abundant
878 compound (Fig. S5). High concentrations of toluene are occasionally found ($> 100 \mu\text{g m}^{-3}$),
879 and benzene is normally about one or two orders of magnitude lower than toluene. It should

880 be noted that waste operation vehicles may also contribute significantly to aromatic emissions
881 (Chiriac et al., 2007; Sadowska-Rociek et al., 2009). Abundant concentrations of aromatics
882 have been detected in heavy vehicle-related areas where gas samples have been taken close to
883 waste dumping and compaction vehicles, or on the roads in and around a landfill (Cheng et al.,
884 2018; Chiriac et al., 2007). This indicates potential emissions of aromatics from the exhausts
885 of landfill operation vehicles, and they should be distinguished from overall emissions when
886 quantifying the fugitive emission of aromatics from landfills.

887

888 ***5.5. Aliphatic hydrocarbons***

889 Saturated and unsaturated aliphatic hydrocarbons are generally considered as xenobiotic
890 compounds and are inherited from disposed waste products. Their probable sources include
891 mixed papers, plastic packaging, fragrant detergents, solvents, refrigerants, waste cooking oil
892 and pesticides on fruits and vegetables (Allen et al., 1997; Gallego et al., 2012; He et al., 2015;
893 Komilis et al., 2004). Aliphatic hydrocarbons are a large chemical group containing various
894 compounds, and they normally make up a significant part of the total concentration (Fig. 6);
895 however, they are seldom given attention in the literature, so it is difficult to summarise
896 common compounds based on the current database.

897 The presence of aliphatic hydrocarbons in raw LFG is largely associated with the age of
898 waste and how far it has decomposed (Fig. 3). Low-molecular weight, low-solubility alkanes
899 (C2-C6) are more abundant during the early phases of waste degradation (Fig. S6) (Ikeguchi
900 and Watanabe, 1991), probably due to the fast decay of easily biodegradable waste (Tan et al.,
901 2017), whereas higher-molecular weight alkanes ($> C6$) tend to dominate the methane
902 production stages (Fig. S2) (Zhang et al., 2012b). The generation of heavy compounds might
903 be ascribed to the degradation of more resistant organic molecules during the progression of
904 waste stabilisation (Chiriac et al., 2011). Nonane, decane, undecane and dodecane have been
905 commonly found in raw LFG, with concentrations ranging from 10^3 to near $10^6 \mu\text{g m}^{-3}$ (Table
906 S1 and Fig. S2). Chiriac et al. (2011) found that alkanes represented about 45% of the total
907 VOCs in raw LFG emitted from old waste cells, and heavier alkanes increased in line with the
908 progression of stabilisation. Decane and its branched isomers represented 42% of the alkanes

909 in their study. It should be mentioned that in this paper, we only summarised a limited number
910 of linear alkanes, but there is also a wide range of different branched alkanes in raw LFG at
911 considerable high concentrations, and excluding them might lead to a bias in the current
912 conclusions (e.g. aliphatic hydrocarbons could be the most abundant chemical group in raw
913 LFG).

914 The concentrations of aliphatic hydrocarbons in ambient air above different landfill units
915 is distributed more evenly, but in general, C2-C6 alkanes are more abundant than heavier
916 alkanes, and the average concentrations for individual compounds are below $100 \mu\text{g m}^{-3}$ (Fig.
917 S6). Higher concentrations of short chain alkanes are primarily detected on the working face
918 (Duan et al., 2014), where waste is in the initial decomposition stage. High concentration
919 levels of alkenes might also appear during this period (Tan et al., 2017), but in most cases
920 they are found in much lower concentrations compared to alkanes (Allen et al., 1997; Parker
921 et al., 2002).

922

923 ***5.6. Halogenated compounds***

924 Halogenated compounds are xenobiotic and derive from the direct volatilisation of
925 compounds contained in dry-cleaning solvent, refrigerants, aerosols, foam-blowing agents,
926 soft foam plastics, paint remover and fire extinguishers (Allen et al., 1997; Hodson, 2008; Liu
927 et al., 2017; Slack et al., 2005; Young and Parker, 1983). Some of the simpler halogenated
928 compounds can be produced from the degradation of higher molecular halogenated
929 compounds; for example, vinyl chloride can be generated through anaerobic
930 hydrogen/halogen substitution of tetrachloroethylene and trichloroethylene in waste (Vogel
931 and McCarty, 1985). Reductive dehalogenation of CFCs and hydrochlorofluorocarbons
932 (HCFCs) can also occur under anaerobic conditions and produce lower molecular halogenated
933 compounds (Scheutz et al., 2010).

934 The concentration of halogenated compounds in raw LFG is mainly governed by waste
935 composition. Compared to other trace gases, halogenated compounds make up a small
936 fraction of the raw LFG (Fig. 6), and the average concentrations for most compounds range
937 from < 1 to $10^4 \mu\text{g m}^{-3}$ (Table S1). Tetrachloromethane, 1,2-dichloroethylene,

938 trichloroethylene, tetrachloroethylene and chlorobenzene are generally more abundant than
939 other compounds in this group (Fig. S2), and the highest concentration in raw LFG has been
940 reported by Tassi et al. (2009) at $5.8 \cdot 10^4 \mu\text{g m}^{-3}$ for tetrachloroethylene. In addition,
941 considerable amounts of CFCs have also been detected in several studies, probably due to the
942 disposal of CFC-containing products (Liu et al., 2017). Vinyl chloride, albeit at relatively
943 lower concentrations in raw LFG (average concentration was $1.9 \cdot 10^3 \mu\text{g m}^{-3}$), might pose a
944 carcinogenic risk to human health when released into the air.

945 Compounds that dominate ambient concentrations above different landfill units are
946 slightly different to those present in raw LFG (Fig. S7). Tetrachloroethylene transpires to have
947 the highest concentration reported by Chiriac et al. (2007), namely $3.7 \cdot 10^3 \mu\text{g m}^{-3}$ measured
948 in the working face area (Table S1). Dichloromethane, 1,2-dichlorobenzene,
949 1,4-dichlorobenzene and CFC-11 have also occasionally been found at high concentrations
950 ranging between 100 and $500 \mu\text{g m}^{-3}$ (Table S1). However, in most cases, the concentration of
951 individual halogenated compounds in ambient landfill air does not exceed $100 \mu\text{g m}^{-3}$, even at
952 the working face (Fig. S7).

953

954 **5.7. Terpenes**

955 Terpenes are derived from both biogenic processes and the direct volatilisation of
956 substances contained in waste. Biogenic sources include plant waste and food materials such
957 as shrubs, grass clippings, woodchips, prunings, vegetables and fruit (Curren et al., 2016;
958 Komilis et al., 2004). Monoterpenes exist widely in vegetation and make an important
959 contribution to plant/fruit fragrance (Lerdau, 1997), and they are released during the
960 degradation of green waste, as well-documented in the literature (Bueyueksoenmez and Evans,
961 2007; Curren et al., 2016; Komilis et al., 2004). In addition, terpenes can also be formed as
962 by-products during the aerobic and anaerobic degradation of lignin (Eitzer, 1995; Pierucci et
963 al., 2005; Wang and Wu, 2008). Other possible sources responsible for terpene emission
964 include flavoured and fragranced household detergents, pharmaceuticals, air fresheners,
965 solvents and resins disposed in MSW landfills (Gallego et al., 2012; Slack et al., 2005).

966 The emission peak of terpenes appears during the initial waste degradation stage,

967 followed by a relatively stationary phase denoted by lower and constant emission rates (Fig. 3)
968 (Canzano et al., 2010; Pierucci et al., 2005; Staley et al., 2006). Their concentrations in raw
969 LFG are associated with waste age, and higher concentrations have been observed within raw
970 LFG in younger landfills. Limonene, which is recognised as a typical tracer of fresh waste,
971 usually dominates the initial waste degradation stage, with a reported highest concentration of
972 $1.9 \cdot 10^5 \mu\text{g m}^{-3}$ in the literature (Table S1) (Canzano et al., 2010; Wang and Wu, 2008). Its
973 concentration decreases as the decomposition process moves forward, and p-cymene
974 gradually takes over to become the major terpene, which is characteristic of old waste, albeit
975 the concentration level is usually lower (Fig. S2) (Davoli et al., 2003; Moreno et al., 2014;
976 Sadowska-Rociek et al., 2009). α -Pinene is also a major compound in the terpene group, with
977 an average concentration level comparable with that of cymene ($4.1 \cdot 10^3 \mu\text{g m}^{-3}$). Allen et al.
978 (1997) stated that an overall percentage of α - and β -pinene above 50% in total terpenes might
979 indicate that the terpene emissions originate from green waste disposed in the landfill.
980 However, this is not always true, since the main terpene compound differs depending on the
981 disposed plant type (Geron et al., 2000). For example, when large amounts of citrus plants are
982 dumped in landfills, there is a significant emission of limonene, thus leading to a low
983 percentage of α - and β -pinene in total terpenes, whilst they still originate from green waste.

984 Concentrations of terpenes measured in ambient air above different landfill units follow
985 the same trend as that found in raw LFG (Fig. S8). Limonene dominates the working face and
986 is associated with fresh waste, and its highest concentration has been identified at $2.2 \cdot 10^3 \mu\text{g}$
987 m^{-3} . In contrast, p-cymene is the leading compound in covered surfaces as well as
988 leachate-related areas typically represented by relatively old waste (Fig. S8). The combination
989 of limonene and p-cymene concentrations can be used as an indicator for a rough estimate of
990 the waste decomposition status, as exhibited well in Table S1 and Fig. S8, in that higher
991 limonene concentrations indicate younger waste at the initial decomposition stage, while
992 increased concentrations of p-cymene appear in older waste.

993

994 **5.8. Nitrogen compounds**

995 Ammonia (NH_3) and amines are produced as a result of the degradation of organic

996 contents in MSW, such as lipid- and protein-containing food and fruit waste, livestock and
997 poultry waste (Fang et al., 2012; Lou et al., 2015). Organic nitrogen contained in this waste
998 can be transferred into ammonia as a result of mineralisation, and this process is more likely
999 to occur under anaerobic conditions or when nitrogen is abundant, i.e. when the C:N ratio of
1000 the organic matter is low (Kissel et al., 1992). Amines, trimethylamine in particular, are an
1001 end product of microbial decomposition that can be produced by the anaerobic breakdown of
1002 amino acids (Wilber and Murray, 1990), and they are often associated with rotten fish or
1003 livestock (Aguirre et al., 2018).

1004 MSW landfills are widely recognised as an important anthropogenic source of NH_3
1005 emissions. According to the European Commission, NH_3 makes up $< 0.1 - 1\%$ of the volume
1006 in biogas produced by anaerobic decomposition (EC, 2006), and the US EPA has suggested
1007 that an $\text{NH}_3:\text{CH}_4$ mass ratio of 0.73% can be used as the emission factor for estimating NH_3
1008 emissions from MSW landfills (Roe et al., 2004). However, very little information can be
1009 found in the literature about NH_3 concentration levels in raw LFG (Table S1). Relatively low
1010 concentrations of NH_3 from passive vents at a Chinese landfill were reported by Fang et al.
1011 (2012) and Cheng et al. (2018), which were $1.5 \cdot 10^3 \mu\text{g m}^{-3}$ and $10.7 \mu\text{g m}^{-3}$, respectively. At
1012 LFG leakage points, much higher NH_3 concentrations were detected by Moreno et al. (2014),
1013 ranging from $1.2 \cdot 10^5$ to $4.8 \cdot 10^5 \mu\text{g m}^{-3}$.

1014 NH_3 concentrations in ambient air above landfill surfaces have been reported at several
1015 sites (Fig. S8). In general, concentrations are very high (averaged concentrations ranging
1016 between 103 and $4.0 \cdot 10^3 \mu\text{g m}^{-3}$) (Table S1 in SM), and in some cases NH_3 is the most
1017 abundant trace gas (Fang et al., 2015; Lim et al., 2018). When the landfill receives sewage
1018 sludge, or has a sewage sludge treatment unit, the ambient NH_3 concentration can be even
1019 higher, reaching up to $5.7 \cdot 10^4 \mu\text{g m}^{-3}$, which significantly exceeds the regulated concentration
1020 (Fang et al., 2012). Leachate collection, storage and treatment areas also tend to have higher
1021 NH_3 concentrations than other parts of a landfill (Fig. S8), since the leachate normally
1022 contains a high level of ammonium (NH_4^+), which can be easily transformed into NH_3 in
1023 above-neutral pH and high-temperature conditions. Trimethylamine is seldom given attention
1024 in the literature although it is an important odorant, and only one ambient concentration

1025 dataset has been reported by Wu et al. (2017): the average concentration of trimethylamine
1026 from six landfill units was 33.4 $\mu\text{g m}^{-3}$ (range 7.7 - 252.9 $\mu\text{g m}^{-3}$), much lower than measured
1027 NH_3 concentrations.

1028

1029 **6. Reported fugitive emission rates of trace gases from different landfill surfaces**

1030

1031 As has been discussed in Section 4, the quantitative measurement of trace gas emission
1032 rates from landfill surfaces is a challenging task, and only a few publications have reported in
1033 this regard. Table 3 summarises the reported emission rates from studies published after 2002,
1034 and most of the measurements were conducted by using surface flux chambers, particularly
1035 static chambers. Detailed concentration data for individual compounds and relevant landfill
1036 information can be found in Table S3 and Table S4.

1037 As evident in Table 3, trace gas emission rates varied by several orders of magnitude at
1038 different landfills. The highest emission rates were found at the active working
1039 area/uncovered waste surface, with total emission rates of all trace gases reaching $\sim 10^8 \mu\text{g m}^{-2}$
1040 d^{-1} (Liu, 2017). Oxygenated compounds, aromatics, aliphatic hydrocarbons and halogenated
1041 compounds turned out to be dominant in the total surface emissions. As discussed in Section 5,
1042 high concentrations of those compounds are often measured in ambient air above active
1043 working area/uncovered waste surfaces, and they are likely to be a result of high emission rate
1044 of those compounds from the relatively fresh waste and underlying older waste.

1045

1046 *****Table 3****

1047 The temporary soil cover in a Chinese landfill showed interestingly higher emission rates
1048 of trace gases than the working face (Han, 2013). This was possibly a result of different waste
1049 age as well as climatic condition during measurement. On the working face, the waste was
1050 relatively fresh (10 days – 2 months), whereas the waste in the covered area was about 2 years
1051 old. It could be expected that more CH₄ was produced by the older waste in the covered area.
1052 In addition, the measurements were conducted during autumn and winter, when the
1053 temperature was low (-1 – 13.7 °C) and CH₄ oxidation in the cover soil was not significant.
1054 Consequently, higher CH₄ emission rates were measured on the covered surface (263 g m⁻² d⁻¹)
1055 than the working face (183 g m⁻² d⁻¹), which indicated an overall higher gas emission from the
1056 covered surface resulting in high trace gas emissions. Schuetz et al. (2003) also reported
1057 relatively high trace gas emission rates from the temporary cover compared to the final cover.
1058 Although the limited data in Table 3 do not support a solid conclusion, it seems that
1059 temporary soil covers are not effective in mitigating trace gas emissions from MSW landfills.
1060 Hence, attention has to be given to temporary soil covers when monitoring trace gas
1061 emissions from MSW landfills.

1062 The emission rates of trace gases from conventional final soil covers are considerably
1063 lower compared to those measured at uncovered waste surfaces and temporary covers, and
1064 negative emission rates have been measured in some studies (Table 3), indicating the
1065 significant reductive oxidation and degradation of trace gases in soil covers. However,
1066 conventional soil covers are not always efficient in reducing trace gas emissions, since cracks
1067 or fissure often develop on soil covers, which results in hotspots characterised by high trace
1068 gas emissions (Scheutz et al., 2008). In addition, different chemical groups have different
1069 degradation processes in the cover soil and some compounds are very difficult to oxidise or
1070 degrade (Barlaz et al., 2004; Han, 2013; Scheutz et al., 2003, 2008). Compounds with higher
1071 degradation rates such as aromatics have low or even negative surface emissions (Schuetz et
1072 al., 2003; 2008), whilst non-degradable compounds such as fully substituted carbons might
1073 have relatively high emission rates at landfill surfaces, as shown in Table 3. In contrast,
1074 installing an HDPE membrane or a biocover in the final cover system seem to be more
1075 effective in significantly reducing fugitive trace gas emissions, as shown in Table 3. albeit

1076 biocovers seem to be less effective in mitigating halogenated compounds (Barlaz et al., 2004;
1077 Scheutz et al., 2008, 2004). Using an HDPE membrane as part of the intermediate/final cover
1078 system has been proven effective in reducing odour pollution from MSW landfills (Ding et al.,
1079 2012), and is widely adopted in Chinese landfills as an odour control technique (Table S2).
1080 However, HDPE membranes easily break and leakage of raw gas from the break and the joint
1081 points will lead to high emission hotspots (Yao et al., 2019). Biocovers also show a great
1082 potential of mitigating odour and trace gas emissions from MSW landfills (e.g. Bogner et al.,
1083 2010; Capanema et al., 2014; Lee et al., 2017), and is more environmentally friendly
1084 considering the recycling of materials (e.g. compost from green waste) and the widely known
1085 strong CH₄ oxidation ability. Hence, more attention should be given on biocovers as a trace
1086 gas emission control technology in future studies.

1087 It is worth mentioning that measurement technique could also influence the results. Table
1088 4 lists trace gas emission rates from the working face of a Chinese landfill, which were
1089 measured by a static chamber and a wind tunnel in a same year. The emission rates measured
1090 by the wind tunnel are in general 2 – 3 orders of magnitude higher than those measured by the
1091 static chamber, and the major chemical groups are also different. Although waste composition
1092 and meteorological conditions might vary during the two measurement campaigns, it is likely
1093 that the different measurement techniques also contributed to the large differences. As
1094 mentioned in Section 4.1.2, it has been found that static chambers normally underestimate the
1095 emission rate, while wind tunnels could overestimate. This might be further enhanced for
1096 measurements on the working face where emissions are usually high, and concentration
1097 built-up develops more quickly inside the static chamber, which suppresses the emission rate.
1098 In contrast, the air flow applied in wind tunnel might lead to a decreased pressure inside and
1099 strips the trace gas from underlying waste below the working face. The different dominant
1100 chemical groups might be attributed to the different analytical technique used in the two
1101 studies. Trace gas samples in the first study were analysed ex-situ by a bench-top GC-MS
1102 system, which has a larger injection volume (400 mL) and longer analysis time (40 min), as
1103 described by Duan et al. (2014); the later study used a portable GC-MS system with a smaller
1104 sample injection volume (100 mL) as well as shorter analysis time (15 min) (Liu et al., 2015).

1105 Since the sensitivity of the portable instrument was lower than the bench-top instrument, less
1106 compounds were detected and bias might exist when comparing the results from the two
1107 studies. For example, the portable GC-MS could not measure ethanol and this compound was
1108 not included in the presented results (Table S3). However, abundant amount of ethanol was
1109 detected in the other study by bench-top instrument, which is consistent with previous study
1110 conducted at the same site (Duan et al., 2014). Hence, one has to be careful when making
1111 comparisons when comparing data from different studies and take into account the specific
1112 methodologies utilised. In particular, further studies are needed to establish the accuracy
1113 range of different emission rate measurement techniques, and to determine the most suitable
1114 technique for measuring fugitive trace gas emissions from various landfill surfaces.

1115

1116 *****Table 4*****

1117

1118 **7. Trace gas dispersion in the atmosphere, and environmental concerns**

1119

1120 To evaluate the environmental impacts and health risks of an existing or planned landfill
1121 site, it is necessary to monitor the dispersion of trace gases in the atmosphere. This is
1122 normally done by using air dispersion models, which include simple or advanced Gaussian
1123 plume models, sophisticated Lagrangian particle models, Eulerian grid models (3-D models)
1124 and the most complicated computational fluid dynamics (CFD) models (Capelli et al., 2013b).
1125 The most widely used options are Gaussian plume models and Lagrangian puff/particle
1126 models (Hayes et al., 2014), such as AERMOD (AMS/EPA Regulatory Model), ADMS
1127 (Atmospheric Dispersion Modelling System,) ISC3 (Industrial Source Complex 3),
1128 CALPUFF (California Puff), AUSTAL, etc. (Capelli et al., 2013b).

1129 Basic input data into these models include meteorological, topographical and trace gas
1130 emission rates (Capelli et al., 2013b). When representative emission rates at a landfill site are
1131 available, the dispersion of trace gases in the atmosphere can be estimated, and downwind
1132 concentrations are obtained for impact assessments of existing landfills, or for predicting
1133 potential impact zones of planned landfills for decision-making processes (e.g. Cremades,

1134 2004; Saral et al., 2009; Demir et al., 2012; Chemel et al., 2012; Abdul-Wahab et al., 2016;
1135 Gallego et al., 2016; Liu et al., 2016a). Detailed descriptions of several cited publications can
1136 be found in the following sections.

1137

1138 ***7.1. Human health risks***

1139 The primary concern related to trace gas emissions from landfills is the health risks they
1140 can pose to on-site workers and residents living in the vicinity following short or long-term
1141 exposure (Giusti, 2009; Lee and Jones-Lee, 1994; Shen et al., 1990). Some compounds that
1142 are usually present in these gases, such as benzene, vinyl chloride and 1,3-butadiene, are
1143 well-known carcinogens, and a number of trace compounds, such as chloroform, carbon
1144 tetrachloride and styrene, are possible or probable carcinogens (IARC, 2010). CFCs also
1145 contribute to non-carcinogenic human toxicities in waste disposal facilities (Zhao et al., 2015).
1146 In addition, many VOCs can cause allergies and asthma and are negatively associated with
1147 lung function (Cakmak et al., 2014; Kim et al., 2013). It is hence necessary to evaluate the
1148 health risk that an MSW landfill site might pose to surrounding populations.

1149 During the 1990s, several epidemiological studies conducted by Goldberg et al. (1995a,
1150 1995b, 1999) at an MSW landfill in Montreal indicated that there was an elevated low birth
1151 weight among children born to women who lived close to the site, and people might face an
1152 excess risk of developing cancers in many organs such as the liver, stomach, kidney, etc.
1153 Similar studies conducted in the UK have also revealed small excess risks of congenital
1154 anomalies and low or very low birth weight in populations living near landfill sites (Elliott et
1155 al., 2001; Palmer et al., 2005). However, these studies failed to provide significant evidence
1156 of a causal link between human illness and exposure to trace gas emissions from landfills, due
1157 to the lack of exposure assessments (Giusti, 2009).

1158 More recently, scholars have started to evaluate the health impacts associated with
1159 landfill trace gas emissions by performing site-specific risk assessments following the
1160 standard procedure, which often includes four steps: hazard identification (usually limited to
1161 carcinogenic and non-carcinogenic risks), hazard characterisation, exposure assessment
1162 (mostly done via using air dispersion models) and risk characterisation (Rovira et al., 2013).

1163 Many studies conducted in developed countries have found that the carcinogenic (R) and
1164 non-carcinogenic (HI) risks caused by trace gas emissions from MSW landfills fall below
1165 regulation values (e.g. Abdul-Wahab et al., 2016; Davoli et al., 2010; Demir et al., 2012;
1166 Martí et al., 2014; Pagé et al., 2007; Palmiotto et al., 2014). This is probably attributed to the
1167 establishment of more stringent regulations on controlling landfill emissions, the development
1168 of landfilling technologies and the implementation of waste management criteria in these
1169 countries.

1170 However, in some countries, people might suffer from increased health risks due to trace
1171 gas emissions from landfills, particularly in nations where there is poor waste source
1172 segregation and improper landfill operations. In the study conducted by Paraskaki and
1173 Lazaridis (2005), the impact zone was 1.5 km away from the investigated landfill, with target
1174 air pollutant concentrations significantly above the WHO reference lifetime exposure health
1175 criteria. Similarly, people living within 1.5 km distance downwind of an MSW landfill in
1176 China were estimated to have significant carcinogenic risks, due to the fugitive aromatic
1177 emissions leaking from the working face (Liu et al., 2016a). The impact distance in this study
1178 was reported to be as far as 4 km away under a worst-case scenario. Considering the high
1179 emission rates of trace gases reported in Section 6, it is not surprising that this landfill pose
1180 higher health risks. In addition, one should be aware that there are still a number of
1181 uncontrolled open dumping sites or landfills under poor or improper management in
1182 developing countries, where waste management is also poor (Khajuria et al., 2010). These
1183 sites might have major emissions that could pose significant risks to surrounding populations,
1184 and quantitative measurements are thereby needed to estimate trace gas emission rates and to
1185 assess health risks.

1186 Occupational health risk caused by trace gas emissions from MSW landfills is also of
1187 great concern. Durmusoglu et al. (2010) investigated the health risk related to BTEX exposure
1188 for workers at a landfill site by performing a stochastic exposure assessment, using
1189 probability distributions in place of point estimations. They concluded that the BTEX
1190 emission did not pose a health threat to workers at the landfill site. Similarly, Cheng et al.
1191 (2018) found that the carcinogenic risk of trace compounds was negligible at a Chinese

1192 landfill, though H₂S might represent a non-carcinogenic risk to on-site workers. However, at
1193 some sites, workers were not so fortunate; for example, Moolla et al. (2013) found that
1194 benzene and toluene emissions from a landfill in South Africa might pose significant
1195 occupational health risks to employees working on site. Other researches have also reported
1196 increased risks, especially under extreme conditions (Wu et al., 2018; Yaghmaien et al., 2019;
1197 Yao et al., 2019). Protective measures for on-site employees are widely used in developed
1198 countries, such as air filters installed in the pressurised cabs of compaction vehicles, masks
1199 and protective suits. These measures can effectively reduce their exposure to trace gases and
1200 thus reduce the associated hazards (Giusti, 2009), and so they should be adopted in countries
1201 with higher emissions (Wu et al., 2018).

1202

1203 ***7.2. Odour pollution***

1204 Odour pollution is another big problem that is generally associated with trace gas
1205 emissions in MSW landfills (Lee and Jones-Lee, 1994). In many cases, it is one of the most
1206 frequent reasons why people complain and exhibit strong opposition to existing landfill sites,
1207 and it has become one of the main barriers to constructing new facilities (Cai et al., 2015).
1208 Exposure to excess odour could cause a variety of undesirable reactions in a population living
1209 in the vicinity of a landfill site, such as mucosal/eye irritation, headache, upper respiratory
1210 symptoms and emotional stress, all of which could significantly lower life quality (Aatamila
1211 et al., 2011; Heaney et al., 2011; Nicell, 2009). Sometimes, landfill odours serve as a tracer of
1212 potential public health harm associated with both known and unknown gaseous emissions
1213 from MSW landfills (Lee and Jones-Lee, 1994). In addition, when an odour exists, people
1214 tend to believe that there is an increasing health hazard (Hayes et al., 2014), which could
1215 result in strong opposition to landfills.

1216 Odour annoyance occurs when the concentration of an odorous compound (i.e. an
1217 odorant) in the air exceeds its odour threshold (Laor et al., 2014). Landfill odours usually
1218 comprise complex mixtures of a large number of volatile compounds (Sarkar and Hobbs,
1219 2002), and sulphur compounds, together with some oxygenated counterparts, are widely
1220 recognised as the most significant odour contributors at landfills (Brosseau and Heitz, 1994;

1221 Fang et al., 2012; Lim et al., 2018; Loizidou and Kapetanos, 1992). For example, DMS,
1222 DMDS, ethanol, limonene and ammonia have been found by many researches as dominating
1223 odorous compounds in landfills (e.g. Ding et al., 2012; Fang et al., 2012; Guo et al., 2017;
1224 Lim et al., 2018; Lu et al., 2015; Tan et al., 2017; Wu et al., 2017; Yue et al., 2014; Zou et al.,
1225 2003). Esters are partially responsible for the “sweeter” odours typical of fresh waste, while
1226 acids are major contributors to the typical smell of leachate (Young and Parker, 1983). In
1227 addition, Aliphatic hydrocarbons might contribute to the odour evolved in waste bins and
1228 long-term odour in landfills (Statheropoulos et al., 2005; Young and Parker, 1983).

1229 Clear knowledge of the trace gas concentrations and emission rates of a landfill site
1230 could help people better understand odour formation and dispersion processes, and these data
1231 are often crucial when studying the odour problem itself (Hayes et al., 2014; Kim, 2016;
1232 Tagaris et al., 2003). In particular, concentration data are important in identifying significant
1233 odour contributors in a mixture of odorants (Curren et al., 2016; Dincer et al., 2006; Lu et al.,
1234 2015; Rosenfeld et al., 2007). The chemical analysis of trace gases also enables the
1235 specification of odour monitoring instruments or the setting up of odour control systems
1236 (Romain et al., 2008). However, it is very challenging to assess odour nuisance based merely
1237 on the chemical concentration of trace gases, due to the additive, antagonistic or synergistic
1238 interaction effects between different odorants (Wu et al., 2015). Olfactory tests are often
1239 required to evaluate odour impacts. Though many researchers have put a great deal of effort
1240 into correlating chemical concentration with olfactometric results through different methods
1241 (e.g. Dincer et al., 2006; Capelli et al., 2008; Lu et al., 2015; Blazy et al., 2015; Guo et al.,
1242 2017; Wu et al., 2017), none of them has succeeded in finding a solid correlation between the
1243 two. A recent study conducted by Wu et al. (2016) attempted to develop a different method to
1244 convert the chemical concentrations of odorants into odour intensity and odour concentrations.
1245 They stated that although the uncertainty of odour thresholds for single odorants restricted the
1246 reliability of the results, the two methods, namely the sum of the odour intensities (SOI) and
1247 the equivalent of odour concentration (EOC), seemed to be promising in predicting odour
1248 pollution by using chemical concentrations of trace gases when olfactometric results are not
1249 available.

1250

1251 ***7.3. Stratospheric ozone depletion and climate change***

1252 MSW landfills are one of the ‘banks’ for the Montreal Protocol-restricted halocarbons
1253 (MPGs), with ozone depletion potential substances contained in discarded materials (e.g.
1254 refrigerators, air conditioners, aerosol cans, blown insulation foam) slowly released into the
1255 atmosphere (Hodson, 2008). These compounds usually take up a significant portion of the
1256 total halocarbons emitted from landfills, with some of them quickly released into the
1257 atmosphere when MPG-containing material is shredded and deposited (Kjeldsen and Scheutz,
1258 2003; Scheutz et al., 2007). The emission process can last for very long periods (from decade
1259 to several hundred years), depending on the size of the shredded waste (Allen et al., 1997;
1260 Kjeldsen and Jensen, 2001).

1261 Hodson et al. (2008, 2010) conducted a study of seven US and nine UK MSW landfills
1262 to determine the contribution of MSW landfills as a source of ozone-depleting substances
1263 (ODS). Their calculations showed that US MSW landfills only emitted less than 1% of the
1264 total US emissions for the four targeted compounds. ODS emissions from UK landfills was a
1265 bit higher, but it was still within 1%-2% of the total emission, except for CFC-12 (6.3%).
1266 Hence, the authors concluded that landfills were only a minor source of lingering ODS
1267 emissions in both the US and UK, and that the majority of current ODS emissions may still
1268 come from equipment still in use. However, in their study, only raw LFG samples were
1269 analysed, and the fugitive emissions of ODS from landfill surfaces were not considered. As
1270 previously mentioned, an LFG collection system is not 100% efficient, and significant
1271 proportions of trace gases are fugitively emitted through landfill surfaces, especially from the
1272 working face. Hence, the use of raw LFG samples for whole-site emission estimation could
1273 lead to a significant underestimation.

1274 A more recent study conducted by Liu et al. (2017) confirmed that the fugitive emission
1275 of halocarbons from landfill working faces was significant, and the annual emission of
1276 CFC-113 from all Chinese landfills took up around 76% of the total national CFC-113
1277 discharges, whilst other targeted compounds were much smaller or negligible. The global
1278 warming potential of fugitive halocarbon made up about 14.4% of landfill methane emissions,

1279 thus indicating a need for more concern about fugitive halocarbon emissions from landfills.
1280 Cheng et al. (2019) also established that the stratospheric ozone depletion and global warming
1281 impacts of a landfill are mainly caused by four halocarbons (HCFC-142b, CFC-11, CFC-12
1282 and carbon tetrachloride). The production of ODS in China was banned in 2010, which was at
1283 least 10 years later than the more developed European countries and the USA, meaning that
1284 many halogen-containing products have not or just reached the end of their lives and will go
1285 into the waste stream, thus resulting in high emissions from landfills. In addition, we should
1286 be aware that such high halocarbon emissions from landfills might also exist in other
1287 countries. CFC-containing materials (such as insulation foams from home appliances) are
1288 expected to go into landfills for a long time in the future, and they will be slowly released into
1289 the atmosphere for decades. Hence, more studies on such emissions are needed, in order to
1290 obtain a full inventory about global halocarbon emissions and to establish emission control
1291 technologies.

1292

1293 **7.4. Air pollution**

1294 It has long been recognised that selected classes of VOCs, such as aliphatic hydrocarbons
1295 and terpenes, can contribute to formation of secondary air pollutants (e.g. ozone,
1296 peroxyacetylnitrate, formaldehyde) and photochemical smog episodes when NO_x and
1297 sunlight exist (Atkinson, 2000) (Atkinson, 2000; Kroll et al., 2006; Shen et al., 1990; Sun et
1298 al., 2016). However, the quantitative importance of trace gas emissions from landfills in
1299 atmospheric chemistry is poorly understood, due to the lack of emission rate data.

1300 Liu et al. (2016) investigated the annual ozone formation and SOA formation potential of
1301 14 dominant aromatics emitted from the working face of a landfill in China. By upscaling
1302 annual figures from the studied landfill to all disposed MSW in the city, they quantified the
1303 total amount of aromatics emissions from all landfills in Beijing. The results showed that
1304 aromatics contributed approximately 0.1% to air pollution compared to other anthropogenic
1305 sources in the city, with toluene, m,p-xylene and 1,3,5-trimethylbenzene being the major
1306 culprits. Another study conducted by Cheng et al. (2019) estimated the environmental impact
1307 of a landfill by using a life cycle assessment model. Photochemical ozone formation was one

1308 of the major environmental impact categories ($6.7 \cdot 10^{-8}$ - $8.6 \cdot 10^{-5}$ personal equivalent, PE),
1309 albeit overall impacts were insignificant, given the low equivalents for all categories. Toluene,
1310 ethyl benzene and benzene were the dominant contributors to photochemical ozone formation
1311 in this case.

1312 Sulphur compounds emitted from landfill sites can also participate in the photochemical
1313 process in ambient air and lead to SO₂ production. According to Shon et al. (2005), the
1314 reduced sulphur compounds, such as dimethyl sulphide, dimethyl disulphide and hydrogen
1315 sulphide, can influence SO₂ production in and around landfills, but their contributions in
1316 terms of observed SO₂ levels are generally insignificant. Song et al. (2007, 2009) confirmed
1317 this conclusion further by conducting both field sampling and modelling. Hence, we can
1318 conclude that sulphur compound emissions from landfills are more important in the
1319 odour-arousing effect and less significant in causing air pollution in landfill environments.

1320 In general, it seems that MSW landfills are insignificant contributors to air pollution in
1321 cities; however, we do need to understand that even though total emissions of trace gas from
1322 landfills are negligible at regional or provincial levels, they might contribute more
1323 significantly in generating highly ambient VOC concentration levels in the surrounding area
1324 (Kim et al., 2008). This effect can be aggravated when there are intense vehicular activities
1325 caused by landfill construction and operation (Yang et al., 2014), and thus the subject
1326 deserves more attention and further investigation.

1327

1328 **8. Conclusions**

1329

1330 In recent years, concern has grown regarding trace gas emissions from municipal solid
1331 waste (MSW) landfills. This paper provides an overview of the origin, transportation and
1332 emission of these gases in landfill environments, the concentration ranges of common trace
1333 gases in raw landfill gas (LFG) and in ambient air above different landfill units, their fugitive
1334 emission fluxes from landfill surfaces and the resulting environmental impacts. In addition,
1335 gas and air sample collection, analytical techniques and quantitative emission measurement
1336 methods for studying trace gas emissions from MSW landfills are presented, and their

1337 advantages and limitations are discussed.

1338 Trace gases in an MSW landfill can originate from the biodegradation of organic waste,
1339 the direct volatilisation of chemicals in waste products or the reactions and conversion of
1340 other trace compounds, all of which undergo various processes inside the waste body before
1341 they are collected and destroyed in gas engines or flares, or fugitively emitted through
1342 installations (vents, leakages in pipes, etc.) and/or from landfill surfaces. Trace gases can be
1343 classified into seven chemical groups according to their chemical structure and functional
1344 units, namely sulphur compounds, oxygenated compounds, aromatics, hydrocarbons, terpenes
1345 and nitrogen compounds. The concentration and presence of trace gases in raw LFG will
1346 depend on the age and stabilisation status of the disposed waste. In general, organic sulphur
1347 and oxygenated compounds are associated with fresh waste, whereas hydrogen sulphide (H₂S),
1348 aromatics and aliphatic hydrocarbon usually appear in high concentrations during the
1349 methanogenesis stage. The concentrations of trace gases in raw LFG are generally three to
1350 four orders of magnitude higher than ambient air concentrations measured above landfill
1351 surfaces. Higher ambient concentrations are mostly observed above uncovered waste cells
1352 with recently disposed waste, and at the working face. Similarly, high emission rates have
1353 been found at uncovered waste surfaces, indicating that fugitive emissions from these areas
1354 are an important source for trace gas emissions in MSW landfills. Final cover systems can
1355 sufficiently reduce the fugitive emission of trace gases from landfills, and utilising
1356 geomembranes in the cover system or installing biocovers are more effective than
1357 conventional soil covers.

1358 Various techniques exist for collecting and analysing trace gases in air or gas samples
1359 obtained from MSW landfills. The most applied sampling techniques are bag sampling and
1360 sorbent tube active sampling, and GC-MS coupled with different pre-concentration steps is
1361 widely used in sample analysis. For quantitative emission measurement, several techniques
1362 have been applied in previous publications, including direct and indirect measurement
1363 techniques. Among them, surface flux chambers, especially static flux chambers, are the most
1364 applied in the literature. The use of different techniques might generate very different results,
1365 even when applied at one site, which has to be taken into consideration when reporting data

1366 and making comparison with previous studies, and so validation testing is necessary. .

1367 Trace gas emissions from MSW landfills might cause increased health risks to
1368 surrounding populations, especially in countries with poor waste source segregation and
1369 landfill operations. However, significant evidence is missing for a causal link between
1370 exposure to trace gas emissions from landfills and human illness. Odour pollution aroused by
1371 trace gas emissions from MSW landfills is widely reported in many countries, and
1372 investigating composition, concentration and emission rates is helpful in predicting odour
1373 pollution, monitoring odour dispersion as well as establishing control measures. Trace gas
1374 emissions from MSW landfills might also contribute to stratospheric ozone depletion, climate
1375 change and regional air pollution; however, their influences are rarely quantitatively
1376 estimated, and so they require further investigation.

1377 Future studies need to establish standardised procedures for the quantitative emission
1378 measurement of trace gases in MSW landfills, and validation is needed to assess the
1379 uncertainties surrounding existing techniques. In addition, a comprehensive emission
1380 inventory is necessary for estimating trace gas emissions from different landfill units, and
1381 particular focus should be given to surface as well as whole-site research. Long-term
1382 monitoring of trace gas emissions from MSW landfills is required to assess further any
1383 potential health risks and to quantify their environmental impacts, particularly in relation to
1384 improperly operated sites in developing countries.

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Tables

Table 1 Overview of the landfill trace gas chemical groups and definitions in this paper.

Chemical group	Definition in this paper
Sulphur compounds	Inorganic and organic sulphur compounds
Oxygenated compounds	Alcohols, ketones, aldehydes, esters and ethers, acetates and acids, furans, volatile fatty acids (VFA), etc.
Aromatics	Compounds containing one or more benzene rings
Aliphatic hydrocarbons	Mainly aliphatic straight, branched or cyclic alkanes (other than methane) and alkenes
Halogenated compounds	Compounds containing F, Cl or Br
Terpenes	A subgroup of hydrocarbon that has a five-carbon isoprene unit; in this paper, we only focus on four monoterpenes – as shown in the Supplementary Material
Nitrogen compounds	N containing compounds; in this paper, we only focus on ammonia and trimethylamine

Table 2 Advantages and limitations of different sampling techniques for the measurement of trace gas concentrations in raw LFG and ambient air.

Sampling technique	Advantages	Limitations
<i>Whole-air sampling techniques</i>		
<i>Sampling bags</i>	<ul style="list-style-type: none"> • cheap; • multiple volumes to choose from; • few requirements for the sampling device. 	<ul style="list-style-type: none"> • potential significant loss or alteration in chemical composition during storage; • limited re-use time; • short storage time (normally < 24h);
<i>Canisters</i>	<ul style="list-style-type: none"> • longer storage time than sampling bags; • both passive and active sampling possible; • reusable many times. 	<ul style="list-style-type: none"> • expensive; • possible analyte loss, due to wall effect, the matrix effect, condensation, etc. • difficult to clean and evacuate.
<i>Glass-bombs</i>	<ul style="list-style-type: none"> • cheap; • inert and easy to clean; • longer storage time than sampling bags. 	<ul style="list-style-type: none"> • fragile for long-term storage and transportation; • possible condensation onto the glass wall; • not compatible with modern analytical instruments.
<i>Sorbent trapping techniques</i>		
<i>Sorbent tubes</i>	<ul style="list-style-type: none"> • long storage time; • various commercially available tubes targeting different compounds; • reusable many times; • increased sensitivity for analysing trace-level compounds; • provide versatile options by combining with other techniques (e.g. underground probes and unmanned aerial vehicles); • possibility of automated-analysis. 	<ul style="list-style-type: none"> • possible loss of certain compounds in the sampled air; • risk of breakthrough on the sorbent bed; • large interference of the analytical procedure when sampling air with high humidity and ozone levels; • possible reaction between sulphur compounds and carbon-based sorbent tubes; • higher initial costs than sampling bags.
<i>Diffusive (passive) samplers</i>	<ul style="list-style-type: none"> • cheap; • small in size; • no need for an air pump or a flow meter; • suitable for long-term sampling (from hours to weeks); • time-averaged concentration can be determined without knowing the sampling volume; • various commercially available choices. 	<ul style="list-style-type: none"> • significant effect of weather conditions on the sampling process; • need to calibrate the actual flow rate even when it is given for commercial tubes; • formation of artefacts during sampling and storage; • risk of back-diffusion on radial diffusive samplers.

Table 3 Fugitive emission rates of trace gases measured at different landfill surfaces ($\mu\text{g m}^{-2} \text{d}^{-1}$)^a.

Reference	(Schuetz et al., 2003)		(Barlaz et al., 2004)		(Schuetz et al., 2008)			(González et al., 2013)			(Liu et al., 2015)	(Liu, 2017; Liu et al., 2017)	(Han, 2013; Yue et al., 2014)	
	Final soil cover	Temporary cover	Biocover (final)	Final soil cover	Final soil cover	Final soil cover - hotspot	Innovative final cover (with HDPE membrane)	Uncovered waste surface; waste age >3 months	Uncovered waste surface; waste age 2-4 weeks	Uncovered waste surface; waste age <1 day	Working face	Working face	working face	Temporary soil cover
Oxygenated compounds								140863	238795	149698	21974112	10355904	21630	102940
Sulphur compounds								1471	1793	2486	1676160	1007424	35275	11140
Aliphatic hydrocarbons	-50	2039			-74	341	60	7860	31658	17498	12479616	36525600	45445	366905
Terpenes								1082	1442	782	777600	6438528	23920	15705
Aromatics	-13	314	-12	21402	-194	8	-16	47844	54670	59494	2108160	20122560	19480	70725
Halogenated compounds	-62	140	123	2253	70	29	-4	22178	9768	12766	1786752	44563392	34860	44175
Nitrogen compounds					0	0	0							
Total	-125	2493	112	23655	-198	378	40	221299	338126	242724	40802400	119013408	180610	611590

^a The concentrations of different chemical groups presented in this table are a sum of the average concentration of individual compounds presented in Table S3.

Table 4 Fugitive surface emission rates of trace gases measured by different techniques at a Chinese landfill ($\text{mg m}^{-2} \text{d}^{-1}$)

Location and Technique	Working face ^a	
	Static chamber	Working face ^b Wind tunnel (sweeping velocity 0.26 m/s)
Chemical group		
Sulphur compounds	38.06	1007
Oxygenated compounds	88.54	10356
Aromatics	2.58	20123
Aliphatic hydrocarbons	11.96	44563
Halogenated compounds	9.99	36526
Terpenes	0.84	6439
Total	152.0	1.2·10⁵

^a (Li, 2015), measured in 2014. ^b (Liu et al., 2018, 2017, 2016b), measured between 2014-2015.

Figures

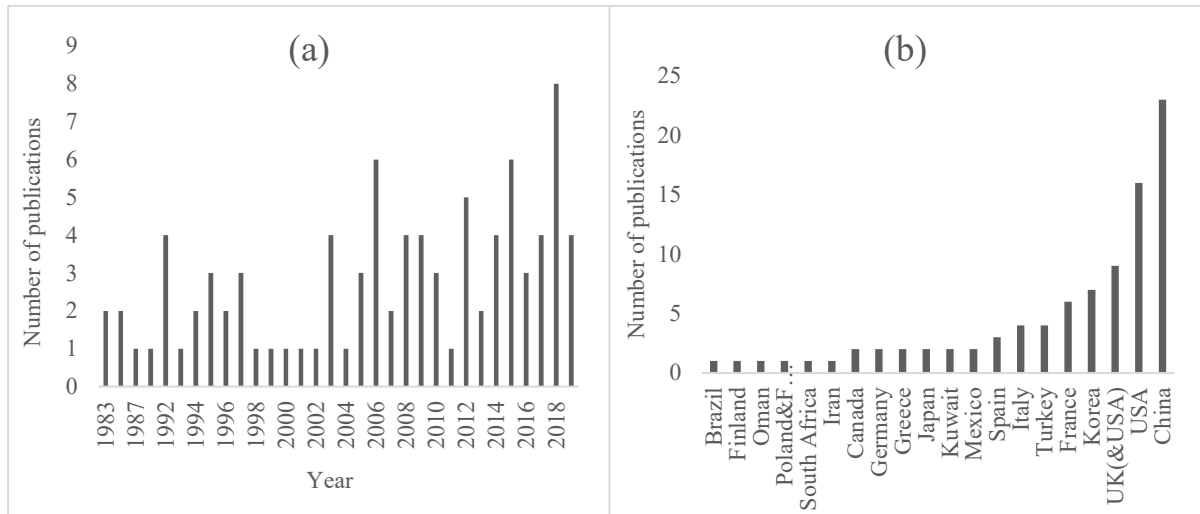


Fig. 1. Overview of the reviewed literature. The countries in Figure 1(b) refer to the location of investigated landfills.

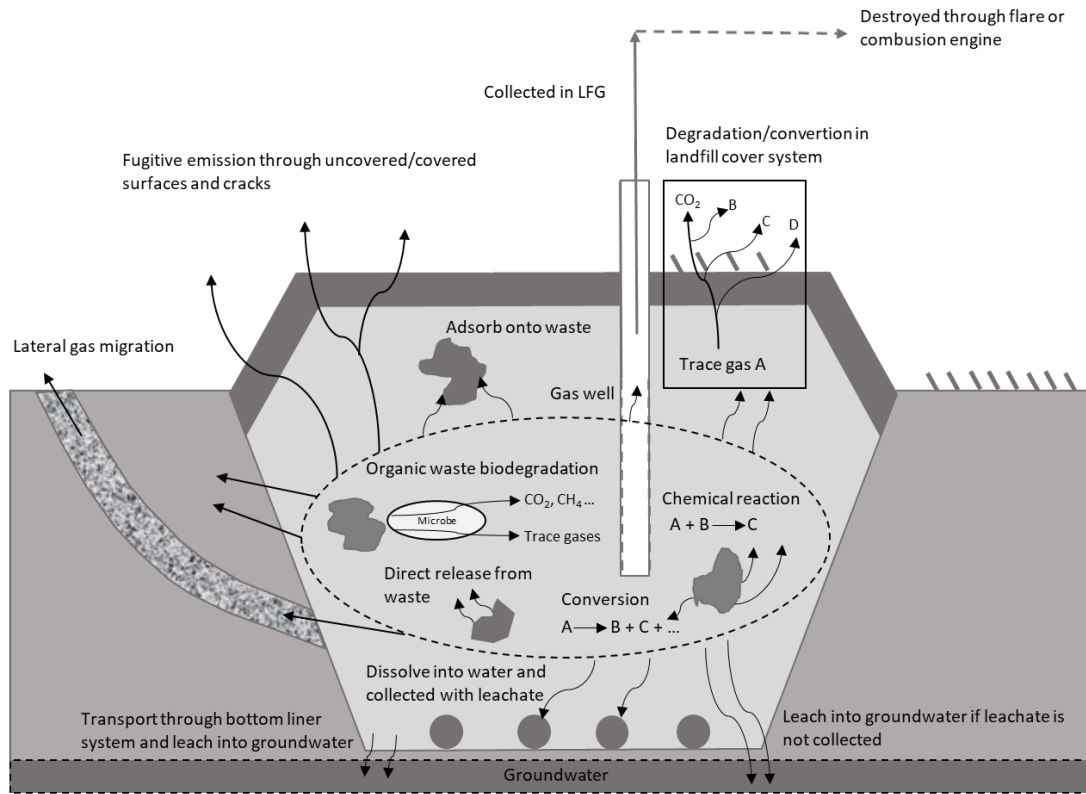


Fig. 2. Trace gas generation, conversion, transportation and emission inside a landfill.

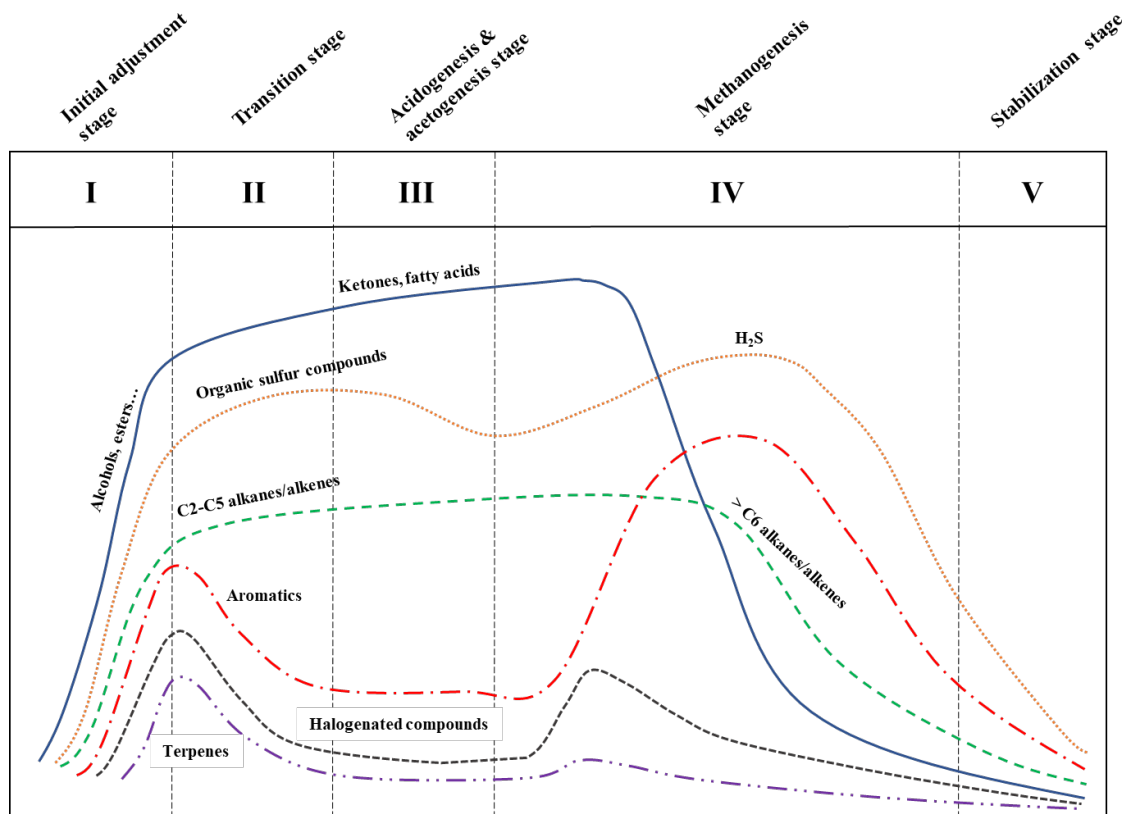


Fig. 3. Trace gas concentration change in raw LFG over time inside a landfill. Nitrogen compounds are not included in this figure due to the limited data available. The figure provides a qualitative assessment of the development of each chemical group in different landfilling stages and might thus not be representative for site-specific situations.

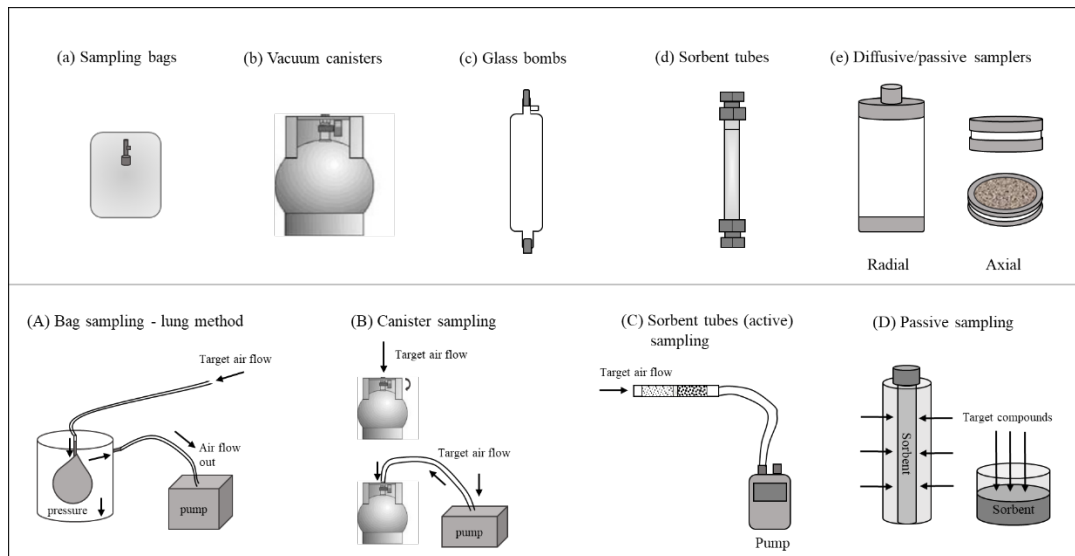


Fig. 4. A schematic overview of sample collection devices and sampling techniques.

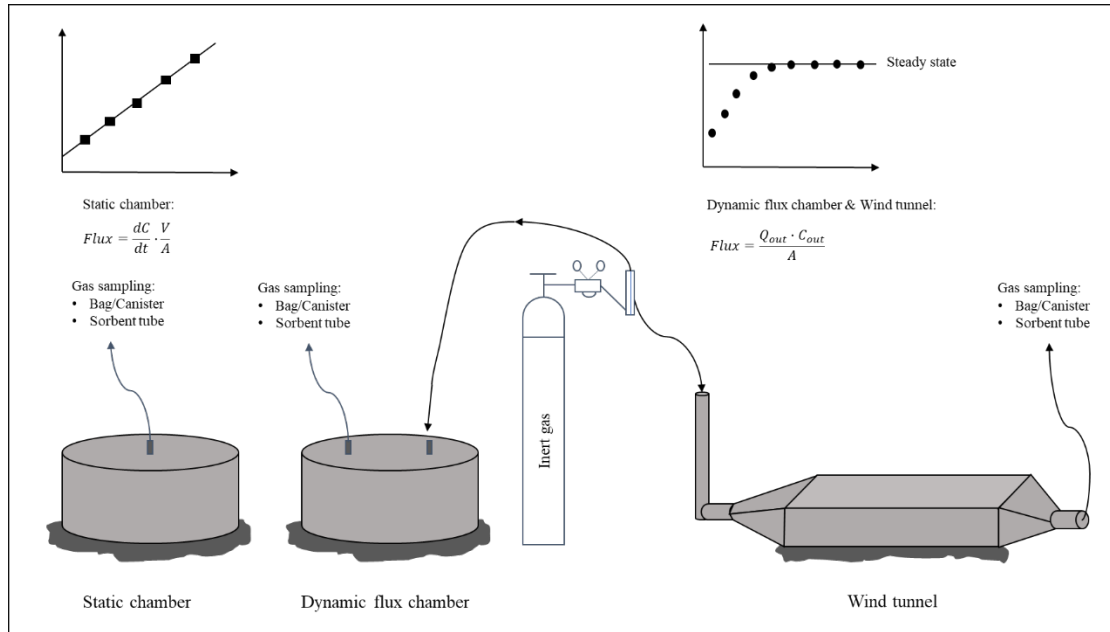


Fig. 5. Schematic overview of surface flux chambers for measuring trace gas emission fluxes from landfill surfaces (Mønster et al., 2019).

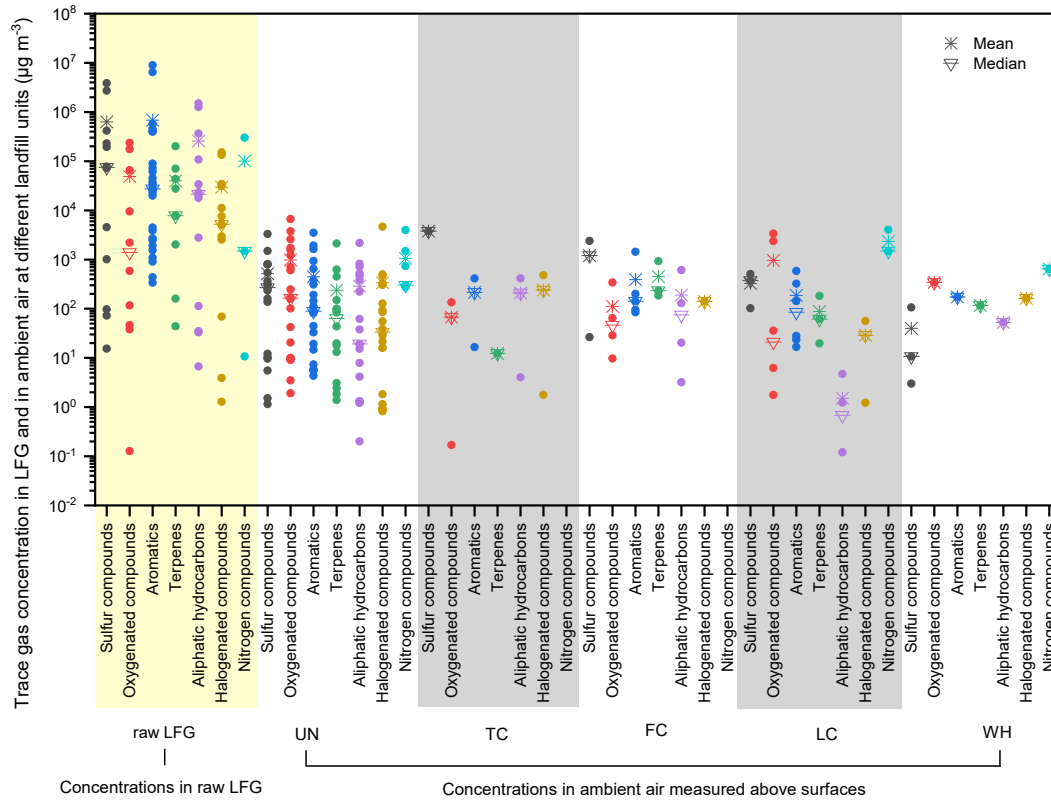


Fig. 6. Trace gas concentrations in LFG and in ambient air at different landfill units, measured above surfaces (UN: active working areas and uncovered surfaces; TC: temporally covered surfaces (intermediate covers); FC: final covers; LC: leachate-related areas; WH: whole-site average). Note: the ‘Not Detected (ND)’ concentrations in Table S1 in SM are not included in this figure; hence, the average values (mean) in the figure are different to those presented in Table S1.