

1 **Testing the performance of one and two box models as tools for risk assessment**
2 **of particle exposure during packing of inorganic fertilizer**

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12 **Highlights:**

- 13 • Occupational exposure to particles during industrial packing was assessed.
14 • No significant increases were found during packing of a granulate fertilizer.
15 • One and two box models predicted adequately actual worker exposure.
16 • Including outdoor concentrations in models was seen to improve their
17 performance.
18 • Models parametrization was seen to be a key issue to adequately predict
19 exposure.

20 **Abstract**

21 Modelling of particle exposure is a useful tool for preliminary exposure assessment in
22 workplaces. However, actual exposure measurements are needed to assess models
23 reliability. Worker exposure was monitored during packing of a complex inorganic
24 granulate fertilizer at industrial scale using small and big bags. Particle concentrations
25 were modelled with one and two box models, where the emission source was
26 estimated with the fertilizer's dustiness index. The exposure levels were used to
27 calculate inhaled dose rates and test accuracy of the exposure modellings. The particle

28 number concentrations were measured from worker area by using a mobility and
29 optical particle sizer which were used to calculate surface area and mass
30 concentrations. The concentrations in the worker area during pre-activity ranged from
31 $63797 - 81073 \text{ cm}^{-3}$, 4.6×10^6 to $7.5 \times 10^6 \text{ um}^2 \text{ cm}^{-3}$, and 354 to $634 \text{ } \mu\text{g m}^{-3}$ (respirable
32 mass fraction) and during packing from 50300 to 85949 cm^{-3} , 4.3×10^6 to $7.6 \times 10^6 \text{ um}^2$
33 cm^{-3} , and 279 to $668 \text{ } \mu\text{g m}^{-3}$ (respirable mass fraction). Thus, the packing process did
34 not significantly increase the exposure levels. High particle number concentration was
35 partly due to the use of diesel-powered forklifts. The particle surface area deposition
36 rate in respiratory tract was up to $7.6 \times 10^6 \text{ } \mu\text{m}^2 \text{ min}^{-1}$ during packing, with 52% - 61% of
37 deposition occurring in the alveolar region. Ratios of the modelled and measured
38 concentrations were 0.98 ± 0.19 and 0.84 ± 0.12 for small and big bags, respectively,
39 when using the one box model, and 0.88 ± 0.25 and 0.82 ± 0.12 , respectively, when
40 using the two box model. The modelling precision improved for both models when
41 outdoor particle concentrations were included. This study shows that exposure
42 concentrations during packing of fertilizers can be predicted with a reasonable
43 accuracy by using a concept of dustiness and mass balance models.

44 **Keywords:** indoor aerosol modelling, exposure prediction, occupational exposure,
45 industrial packing, risk management.

46 **1. Introduction**

47 Industrial bag filling, packing and pouring processes have been pointed out as activities
48 with high potential to emit airborne particles. Studies in different industrial sectors had
49 reported high levels of worker exposure to particles, e.g; during pouring and packing of
50 paint pigments, packing of TiO_2 , carbon black, fullerenes and carbon nanofibres (Ding
51 et al., 2017; Fujitani et al., 2008; Koivisto et al., 2015, 2012a; Koponen et al., 2015;
52 Kuhlbusch et al., 2004, Evans et al., 2010) as well as packing and pouring of cement
53 materials (Notø et al., 2018; Peters et al., 2008). Additionally, differences in particle

54 release have been observed when pouring different materials, different amounts, and
55 using different types of mixing tanks (Koponen et al., 2015). Thus, every case is
56 specific and further research is needed in order to understand emission patterns during
57 packing and pouring.

58 Exposure to particulate matter (PM) is known to cause various adverse health effects,
59 such as pulmonary and cardiovascular diseases and cancer (Landrigan et al., 2017).
60 Current epidemiological and toxicological studies consider PM_{2.5} (with aerodynamic
61 particle diameter $D_p \leq 2.5 \mu\text{m}$) as the most harmful component for human health
62 (Gakidou et al., 2017; Landrigan et al., 2017; World Health Organization, 2016).
63 Inorganic complex fertilizers have been found to be moderately toxic to earthworms
64 (Shruthi et al., 2017). In humans, due to inhalation of fertilizer degradation products,
65 health effects might come up especially after long term exposures (Yara Iberian S.A,
66 2005). Ammonium nitrate, used in complex inorganic fertilizers, when inhaled, was
67 seen to cause possibly meaningful pulmonary function changes (Kleinman et al., 1980)
68 and to be irritating, cause coughing, bronchospasm, laryngospasm and laryngeal
69 edema even at low concentrations (Gorguner and Akgun, 2003). Additionally, the
70 clinical examination of workers of the ammonium nitrate production showed frequent
71 cases of chronic bronchitis and radiculoneuropathy (Tsimakuridze et al., 2005). On the
72 other hand, ammonium nitrate is known to be potentially explosive when confined.
73 Potassium nitrate, also included in some inorganic fertilizers composition, has been
74 seen to be irritating for the respiratory tract (INCHEM, 2001). Therefore, the study of
75 packing of an inorganic fertilizer is of interest as workers can be exposed to relatively
76 high concentrations of airborne fertilizer particles, which might cause respiratory health
77 effects.

78

79 Exposure prediction models have been proposed as valuable risk assessment tools.
80 Since the initial application of exposure prediction models, several research papers

81 have been published regarding their theoretical aspects (Ganser and Hewett, 2017;
82 Hewett and Ganser, 2017; Hussein and Kulmala, 2008; Nazaroff, 2004; Nazaroff and
83 Cass, 1989). The two box model is a well-accepted exposure assessment tool in the
84 risk assessment field as, even with its simplified assumptions, it is able to adequately
85 simulate actual conditions for various processes including volatile compounds and PM
86 emissions (Arnold et al., 2017; Jayjock et al., 2011). In the chemical industry, models
87 have been tested in a variety of cases (Nicas, 2016; Sahmel et al., 2009 and
88 references therein). However, when testing the models for PM in actual industrial
89 environments, the number of studies decreases (Arnold et al., 2017; Boelter et al.,
90 2009; Johnson et al., 2011; Jones et al., 2011; Koivisto et al., 2015; Lopez et al., 2015).
91 Recently, Arnold et al. (2017) conducted a study where the one and two box models,
92 were evaluated under highly controlled conditions. Predicted exposure results for three
93 industrial solvents when using near and far field models was categorized excellent and
94 good to excellent under the ASTM Standard 5157 criteria (Arnold et al., 2017).
95 However, in order to implement prediction models as trustable tools for worker risk
96 assessment, additional real-world cases need to be evaluated to test model
97 performance and to understand the uncertainties related to critical parameters, such as
98 the source characterization, local controls, and air mixing (Jayjock et al., 2011; Sahmel
99 et al., 2009).

100 The objectives of the present study were 1) to perform a worker exposure and risk
101 assessment study of packing of an inorganic complex fertilizer in an industrial plant,
102 and 2) to test the one box and two box models performance in real-world settings in
103 order to contribute to the better understanding and validation of exposure prediction
104 models.

105

106

107 **2. Methodology**

108 **2.1. Work environment and packing process**

109 The measurements were carried out during packing of a fertilizer in two different
110 packing lines between the 23th and 26th of January 2017 at an industrial facility
111 located in Castellón, Spain. The fertilizer (YaraMila COMPLEX, PF595P, YaraIberian
112 S.A.) main components were ammonium nitrate; NH_4NO_3 (15 - 20%), potassium
113 nitrate; KNO_3 (12.5 - 15%) and calcium fluoride; CaF_2 (2 - 3%). The fertilizer was
114 granulated in 2.5 to 5 mm diameter spherical pellets.

115 The packing hall was only naturally ventilated and the replacement air came from
116 outdoors and from adjacent industrial hall via doors, which were always open (Figure
117 1). The packing lines were for small bags (25 kg) and big bags (600 kg) where the
118 studied fertilizer was poured into the bags from a silo by using a feed funnel. Figure S1
119 in the Supporting Information shows photos from the packing lines. The two packing
120 lines were not operated at the same time. Two-day measurements were conducted at
121 both packing lines, small bags day 1 (SB1), small bags day 2 (SB2), big bags day 1
122 (BB1) and big bags day 2 (BB2). In small bags, packing was carried out through an
123 opening fitting the bag width (33-35 cm) and subsequently mechanically sealed. The
124 fertilizer was poured at a flow of 250 kg min^{-1} and the drop height was 5 cm from the
125 feed funnel to the bag opening. Total material drop height was approximately 0.6 m.
126 The packing process was fully automated and the process area was partially enclosed.
127 In big bags, packing was carried out through a cylindrical opening (20 cm diameter)
128 and at a 175 kg min^{-1} flow; material drop height was 20 cm from the feed funnel to the
129 bag opening. Total material drop height was approximately 1.3 m. In that case, the bag
130 was manually closed by the worker, who was standing in front of the bag at
131 approximately 0.5 m distance.

132 During small and big bags filling, workers tasks were to control and guarantee the
133 correct functioning of the lines as well as to move the filled bags to the storage area
134 using an electric forklift. Occasionally, diesel-powered forklifts were performing truck
135 loading and unloading operations in the hall.

136 **2.2. Aerosol measurements and sampling**

137 Particle number and mass concentrations were monitored in real time in the worker
138 area (WA), indoors, and outdoors (Figure 1). All online instruments were synchronized
139 prior to the measurements and intercompared overnight between experiments. Particle
140 concentrations during packing were measured for approximately two hours.
141 Additionally, 30 minutes of pre-activity concentrations were measured for each day
142 except for BB2.

143 In the worker are, the instruments were placed on a portable table at approximately 1
144 m height (instrument inlets being at 1.5 m above the ground level), at 0.5 m from the
145 emission source and 1 m from the worker (Figure 1 and Figure S1, Supporting
146 Information). The monitoring instruments were:

- 147 - An electrical mobility spectrometer (NanoScan, SMPS TSI Model 3910; sample
148 flow rate 0.7 l min^{-1}) to measure particle number concentration and particle size
149 distribution in 13 channels from 10 to 420 nm with a 1 minute time resolution
- 150 - A Mini Wide Range Aerosol Spectrometer (Mini-WRAS 1371; sample flow rate
151 1.2 l min^{-1}) to measure particle mass concentration, particle number
152 concentration and particle size distribution from 10 nm to 35 μm in 41 channels
153 with a 1 minute time resolution
- 154 - A miniature diffusion size classifier (DiSCmini Matter Aerosol, Testo; sample
155 flow rate 1 l min^{-1}) to measure particle number concentration, mean particle size
156 and alveolar lung deposition surface area (LDSA) in a range of 10 to 700 nm
157 with a 1 minute time resolution

158 - A Mini Laser Aerosol Spectrometer (Grimm, Mini-LAS 11R; sample flow rate 1.2
159 l min⁻¹) to measure particle mass concentration from 0.25 to 32 µm in 31
160 channels with a 1 minute time resolution.

161 The indoor and outdoor concentrations were monitored by using a DiSCmini and a
162 Grimm Mini-LAS, with the same settings as described above.

163 During the packing process, particles emitted were collected onto Au grids (Quantifoil
164 ® with 1 µm diameter holes – 4 µm separation of 200 mesh). The grids were attached
165 to polycarbonate filters that were placed in a sampling cassette (SKC INC., USA, inlet
166 diameter 1/8 in. and filter diameter 25 mm). The cassette was connected to a Leland
167 pump with an operating flow rate of 3 l min⁻¹. The morphology and primary particle size
168 of the particles collected were determined using a transmission electron microscope
169 (TEM, Jeol, JEM 1220, Tokyo, Japan) coupled with an energy-dispersive X-ray (EDX)
170 spectrometer.

171 The worker area particle number size distributions measured by the NanoScan and
172 MiniWras were combined according to Koivisto et al. (2012a) to obtain a wide range for
173 particle size distribution from 10 nm to 35 µm. NanoScan size channels between 11.5 -
174 86.6 nm were used while channels ranging from 139 nm to 35 µm were taken from the
175 MiniWras. Between 86.6 nm and 139 nm a combined channel (108.6 nm) was created.
176 Upper channels from NanoScan (> 115.5 nm) were not used as it is known to not have
177 a good resolution for particles >200 nm (Fonseca et al., 2016), while MiniWras was
178 seen to not accurately measure particles under 50 nm; therefore, MiniWras lower
179 channels were not used (see Figure S2, Supporting information) and explanation.
180 Here, due to channels cut, ultrafine particles are defined as $D_p < 86.60$ nm, fine
181 particles as 86.60 nm < $D_p < 943.0$ nm and coarse particles as > 943.0 nm.

182 Increases and reductions in exposure during packing when comparing with pre-activity
183 levels were considered statistically significant when the following approach (Asbach et
184 al., 2012; Kaminski et al., 2015) was fulfilled:

185

186 Mean concentration during packing $> BG \pm 3*(\sigma BG)$

187

188 where BG is the mean temporal background (pre-activity) concentration and σBG is the
189 standard deviation of the background concentration.

190 **2.3. Dustiness**

191 Material dustiness was assessed by using the continuous drop standard method (EN
192 15051). The continuous drop device, made of stainless steel, consisted of a cylindrical
193 pipe through which air circulated in an upward direction with a volume flow rate of 53 l
194 min^{-1} . Concentric to the cylindrical pipe there was an inner pipe, slightly shorter than
195 the cylindrical pipe, through which material was dropped at a flow rate of 6 to 10 g min^{-1} ,
196 so that the powdered material was released into a counter-current airflow (López-
197 Lilao et al., 2015). Total material drop height during the test is approximately 1.2 m.
198 Two sampling heads for inhalable (approximately PST; designed by Institut für
199 Gefahrstoff-Forschung-IGF) and respirable (approximately PM_{10} ; FSP-2, BGIA) fractions
200 were located slightly above the discharge position of the material. Samples for
201 gravimetric measurements of inhalable and respirable fractions were collected on
202 cellulose thimbles, single thickness, 10x50 mm 25/pk and PVC filters of 37 mm and 5
203 μm of porosity respectively. The experiment, which lasted for 10 minutes, was repeated
204 two times to ensure results repeatability. Between experiment repetitions, the sampling
205 heads for inhalable and respirable fractions were superficially cleaned while the rest of
206 the device was thoroughly cleaned only at the end of the test.

207

208 **2.4. Exposure modelling**

209 **2.4.1. Dispersion models**

210 Exposure modelling was performed by using a one box model (Hewett and Ganser,
211 2017) and a two box model (Ganser and Hewett, 2017). Figure 2 shows the models
212 schemes and the mass balance equations. The models assume that 1) particles are
213 fully mixed at all times; 2) mass is created by a source inside the plant (near field in two
214 box model) and by concentrations coming from outdoors; 3) there are no other particle
215 losses than the natural ventilation. The models were used to calculate the respirable
216 fraction. Particle losses by sedimentation may be considered negligible for this size
217 fraction.

218 **2.4.2. Emission source**

219 The emission (S) from the packing process is described based on the dustiness index
220 as:

$$221 \quad S(t) = DI \cdot H \cdot \frac{dM(t)}{dt} \cdot LC \quad (1)$$

222 where DI is the respirable dustiness index of the fertilizer expressed in mg kg^{-1} or
223 particles kg^{-1} , H is the handling energy factor for the process, dM/dt (kg min^{-1}) is the
224 mass flow of the fertilizer, and LC is the protection factor of localized controls. The
225 respirable dustiness index of the fertilizer was obtained using the continuous drop
226 method, as it is the method that adapts better to the process under study (Pujara,
227 1997; Ribalta et al., 2018 under review).

228 **2.4.3. Modelling parametrization**

229 The input parameters needed to run the model and experimentally unavailable in this
230 case study are the handling energy factor (H), local control factors (LC), and the air
231 flow rate (β) between near field (NF) and far field (FF) (for two box model only).

232 By definition, H , links the energy applied during the process with the energy applied
233 during the dustiness test and can range from 0 to 1 (Koivisto et al., 2015; Lidén, 2006;
234 Schneider and Jensen, 2007). Here, H was set to 0.5 for small bags because the drop
235 height during small bags packing was ca. half of the drop height in dustiness test. For
236 big bags, H was assumed to be 1 as material drop height was similar to dustiness drop
237 height (see 2.1 and 2.4). With regard to local controls (LC), two main controls were
238 detected. For both small and big bags, the bag itself was estimated in this work to
239 reduce particle release by 40% (applied in the emission rate equation as $(LC_{\text{bag}} = 0.6)$.
240 In addition, for small bags one box model, the effect of the enclosure was taken into
241 account and applied in the model reducing emission by 50% ($LC_{\text{enclosed}} = 0.5$)
242 (Fransman et al., 2008). Finally, β was estimated after testing the range values
243 reported by Baldwin and Maynard (1998) and Arnold et al. (2017) taking into account
244 the characteristics from our case scenarios. A sensitivity analysis for different β was
245 carried out and is reported in the section below (Table 1). For small bags it was set at
246 $0.75 \text{ m}^3 \text{ min}^{-1}$ (0.0125 m s^{-1}) as the air flow rate was considered to be low due to the
247 enclosure of the packing line (enclosure opening of 1 m^2). In this case, for the two box
248 model, as the effect of the enclosure was introduced by the NF-FF β , the local control
249 regarding the enclosure (LC_{enclosed}) in the emission rate equation was suppressed. For
250 big bags, the air flow rate was considered to be higher as there was no enclosure or
251 division between NF and FF, so β was set to $30 \text{ m}^3 \text{ min}^{-1}$ (0.04 m s^{-1}). The model
252 schemes and parameters are listed in Figure 2. The air exchange rate (AER) between
253 indoor and outdoor air was experimentally calculated considering outdoor wind speed
254 during the measurement period (obtained from the local air quality monitoring network),
255 the size of the outdoors door, and the size of the industrial unit. This resulted in a mean
256 air exchange rate of around 7 h^{-1} for the entire period.

257

258

259 **2.5. Calculated active surface area and mass concentrations**

260 The particle active surface area was calculated by applying particle size distribution
261 obtained from NanoScan and MiniWras data combination to the equation (2) described
262 in Heitbrink et al. (2009) as in Koivisto et al. (2012b).

263
$$S = \frac{3\pi\lambda D_b}{C_c(D_b)\delta} \quad (2)$$

264 where λ is the mean free path for air, 0.066 μm , and δ is the scattering parameter for
265 air, 0.905. D_b is the mobility diameter and C_c the slip correction factor for the
266 corresponding aerodynamic or mobility particle size.

267 The particle mass was additionally calculated by using mobility particle diameter and
268 effective density as in Koivisto et al. (2012b)

269
$$m = \rho_{eff} \frac{\pi}{6} D_b^3 \quad (3)$$

270 where ρ_{eff} is the effective density. As particles density was unknown, 1 g cm^{-3} was
271 assumed for simplicity.

272 **2.6 Calculated regional inhalation dose rate**

273 The inhalation dose of deposited particles in the respiratory system during inspiration
274 and expiration was quantified. The regional inhalation dose rate was obtained by
275 multiplying particle size concentrations on the worker area (NanoScan and MiniWras
276 data combination) by the ICRP human respiratory tract model deposition probability
277 (ICRP, 2011). The respiratory volume used was 25 l min^{-1} , corresponding to male
278 respiration during light exercise (Koivisto et al., 2012b). The regional dose was
279 calculated for head airways, tracheobronchial and alveolar regions by using simplified
280 deposition fraction equations for the ICRP model as described by Hinds (1999). In the

281 model particles were assumed to be spherical and to preserve their size during
282 inhalation.

283 **3. Results**

284 **3.1. Material morphology and characterization**

285 Samples collected onto Au TEM grids were observed and characterized using TEM-
286 EDX. In the samples collected during SB2 (Figure 3a, 3b, 3c, 3d and 3e) and
287 BB1(Figure 3f, 3g, 3h and 3i) experiments, particles which main elements were O, Na,
288 K, Ca, Cr and Zn were detected proving the presence of fertilizer particles in the worker
289 area (Figure 3a, 3b, 3c, 3d 3f, 3g and 3h). A few differences were observed between
290 both samples. Fertilizer particles size was between 1 μm up to > 35 μm in both
291 samples, although in BB1 there was a bigger proportion of bigger particles (Figure 3f)
292 whereas in SB2 a bigger proportion of smaller ones (Figure 3c). Additionally,
293 agglomerates of nanoparticles, with particle size < 50nm and main components O and
294 C, were found on both samples indicating the presence in the worker area of diesel
295 combustible particles, coming from the diesel forklift (Figure 3d, 3e, 3h and 3i). Those
296 agglomerates were occasionally seen in the BB1 samples (Figure 3h and 3i), whereas
297 in the SB2 they were highly abundant (Figure 3c, 3d and 3e) owing to a higher activity
298 of diesel forklifts inside the plant (96.2%; Table 2).

299 **3.2. Dustiness indices**

300 Material dustiness was assessed using the continuous drop method and results were
301 given in terms of inhalable and respirable mass fractions (mg kg^{-1}) gravimetrically
302 analyzed. Following the EN 15051 dustiness classification for continuous drop, the
303 fertilizer under study was classified as a material with low and very low dustiness
304 indices, with 1026 mg kg^{-1} and 16 mg kg^{-1} for inhalable and respirable fractions,
305 respectively.

306 3.3. Concentrations

307 3.3.1. Worker area concentrations

308 The measurements started 34 to 46 minutes prior to the packing process. Packing
309 lasted between 1 h 20 minutes and 2 h 43 minutes (Table 2). For BB2 no background
310 concentrations could be recorded. During SB1 (Figure S3), total particle number and
311 inhalable mass concentrations during packing were similar to background
312 concentrations (Table 3 and Figure S3). Concentrations of fine particles (100 nm - 1
313 μm) and thoracic and respirable mass concentrations were lower during packing
314 compared with pre-activity levels (Table 3, Figure S3, Supporting information), which
315 resulted from decreasing background concentrations during the pre-activity period (see
316 Figure S3). Thus, it was concluded that during SB1 experiments no significant impacts
317 on particle exposure were detected. Similarly, during SB2 (Figure 4) experiments no
318 statistically significant differences were observed in terms of mass concentrations
319 between the pre-activity and activity periods (Table 3, Figure 4). These results are in
320 agreement with the low dustiness index of the fertilizer material. Conversely, during
321 SB2 total particle number concentration did increase significantly with regard to pre-
322 activity levels (on average for total particle number, 17340 cm^{-3}) (Table 3, Figure 4).
323 This increase may have been linked to diesel emissions from a diesel forklift which
324 operated inside the plant during this period, as will be discussed below. In addition,
325 very few differences were observed in particle size distributions between the pre-
326 activity and activity particle size distributions for SB1 and SB2 (Figure 5a and 5b). In
327 Koivisto et al. (2012a) measurements during packing of TiO_2 into small and large bags
328 did not have an impact on particle concentrations except when opening the enclosed
329 packing machine. Impacts on worker exposure when packing silicon nanoparticles
330 were also not detected probably because the packing line was hermetically sealed
331 (Wang et al., 2012).

332 During BB1 (Figure 6), particle number and mass concentrations were again similar to
333 pre-activity concentrations, showing slightly higher (non statistically significant) mean
334 concentrations (Table 3). Total particle number concentrations increased by 4876 cm^{-3}
335 and respirable mass concentration by $314 \mu\text{g m}^{-3}$ (Table 3, Figure 6). During the BB2
336 (Figure S4) experiments pre-activity concentrations were not available because the
337 activity was initiated before the monitoring instrumentation was ready, and therefore
338 worker exposure can only be discussed comparing with indoor background
339 concentrations. As in the case of SB1 and SB2 very few differences were observed in
340 particle size distribution between the pre-activity and BB1 packing periods. Only slight
341 increases in particles $< 30 \text{ nm}$ and $> 10 \mu\text{m}$ were observed (Figures 5c). Contrarily, in
342 Koivisto et al. (2012a), packing of TiO_2 into large bags was seen to increase particles $>$
343 500 nm . Even so, the present results were to be expected as when classifying the
344 fertilizer according to its dustiness index, it was sorted as a material with very low and
345 low capacity to generate airborne dust for inhalable and respirable fractions,
346 respectively.

347 As described above, particle number concentrations increased significantly only during
348 two of the four experiments, i.e., SB2 and BB1. However, those increases were not
349 clearly related to the packing activity as no specific relation was seen with the start and
350 stop of the process (Figures 4 and 6). Increases of ultrafine particles in comparison
351 with the background were always below 40000 cm^{-3} , the suggested reference limit
352 value in this specific case (non-biodegradable granular nanomaterials in the range of
353 $1\text{--}100 \text{ nm}$ and density $< 6 \text{ kg l}^{-1}$) (Van Broekhuizen et al., 2012).

354 Inhalable and respirable mass concentrations did not exceed in any case the limit
355 values for particles not otherwise specified of 10 and 3 mg m^{-3} , respectively (INSH,
356 2018). Thus, it may be concluded that packing activity of the specific fertilizer did not
357 have a significant impact on worker exposure with regard to particles in the $11.5 \text{ nm} -$
358 $35 \mu\text{m}$ size range. It should be pointed out that in this study worker exposure

359 concentrations do not correspond strictly to the worker breathing zone (because
360 instruments were not worn by the workers), which are expected to be higher (Koivisto
361 et al., 2015; Koponen et al., 2015). Additionally, the measurements were carried out for
362 a maximum of 2.5 hours and therefore not representative of the 8 hours necessary to
363 calculate the 8 hr time weighted average over a full shift.

364 Packing processes and similar industrial activities such as material pouring have been
365 previously studied among different types of industries with results indicating that
366 packing, pouring or dumping processes usually lead to slight increases in worker
367 exposure concentrations. Packing of carbon black in bags of 25 kg and 1000 kg was
368 shown to increase airborne particles > 400 nm and mass concentrations (Ding et al.,
369 2017; Kuhlbusch et al., 2004). Fullerenes packing increased particle number > 1000
370 nm (Fujitani et al., 2008). Evans et al. (2010) also found that dumping of carbon
371 nanofibers into a drum resulted in an increase of respirable mass concentrations. In the
372 case of the cement industry, Notø et al. (2018) found that packing was associated with
373 an increase of worker exposure to the thoracic mass fraction of 12% and 33% when
374 working less than and more than half a shift, respectively. On the contrary, pouring of
375 cement at a construction site was seen to have highly variable and low percentages of
376 inhalable mass exposure, probably because of workers performing pouring operations
377 also carried out other activities (Peters et al., 2008). In comparison to these studies, the
378 fertilizer packing case presented in this work seemed to have one of the lowest impacts
379 on worker exposure to particle mass and number concentrations.

380 **3.3.2. Outdoor concentrations**

381 The packing hall was connected by two doors (Figure 1) to outdoors and to another
382 industrial unit. In both sites other processes were occasionally ongoing. Thus, influence
383 of outdoors and other processes taking place in the adjacent industrial unit were to be
384 expected. Outdoor particle number concentrations as well as PM₁₀ mass were usually

385 lower or in a similar range as the worker area and indoor concentrations (thoracic mass
386 fraction) (Table S1 and S2, Supporting information). Regarding mean particle size, it
387 was usually smaller in the outdoor location than in the indoor and worker area by 10 -
388 20 nm (Table S1, Supporting information) due to the influence of outdoor traffic
389 emissions. Mean particle size remained more or less constant between pre-activity and
390 packing periods in the worker area (38-32, 28-37, 33-37, 41-44 nm), indoor (43-37, 38-
391 43 nm) and outdoor (23-20, 31-31, 29-32 nm) measurement points for all days. In
392 general, outdoor concentrations seemed to follow a different pattern from the rest of the
393 locations even if with some exceptions where similar peaks in outdoor, indoor and
394 worker area were observed (e.g., Figure 4, 11:30; Figure S3, 15:10; Figure S4, 12:15).
395 Numerous studies have reported the infiltration of outdoor particles into diverse indoor
396 environments, especially through windows and doors when they are open (Bennett and
397 Koutrakis, 2006; Hussein et al., 2009; Koponen et al., 2001; Reche et al., 2014; Rivas
398 et al., 2015; Wang et al., 2010). In Wang et al. (2010), outdoor infiltration was detected
399 in a similar packing industrial unit where indoor and outdoor areas were connected by
400 opened doors as in the present study.

401 **3.3.3. Forklifts activity**

402 Electric and diesel forklift activity was recorded and is shown on the top of Figures 4, 6,
403 S3 and S4 and as a percentage of total recorded time in Table 2.

404 During the SB1 packing period, an increase in particle number concentration (< 50 nm)
405 was detected when the diesel forklift was driving inside the hall (> 15:00 h) (Figure S3a
406 and S3b). During SB2, only a slight increase in number concentration was observed
407 when the diesel forklift was driving and the electric forklift ended its activity (Figure 4a
408 and 4b, 11:10). During BB1, a slightly increase of particle number concentration (mean
409 size 30 nm) was observed coinciding with the start of a diesel forklift at 10:20 (Figure
410 6a and 6b). During BB2 (Figure S4), two increments of number concentration were

411 detected, but only the first one could be clearly linked to a diesel forklift activity. On
412 some occasions, increases in particle number concentrations in the worker area and
413 indoor seemed to be related to the use of the diesel forklift while in others this
414 relationship was more difficult to establish. For example, the highest statistically
415 significant increase in mean particle number concentration in the worker area was for
416 SB2, also having the highest percentage of diesel forklift activity 96.2% (Table 2).
417 Moreover, when an increase in number concentration linked to the activity of a diesel
418 forklift was seen in the worker area it was also seen in the outdoor and indoor
419 measurement points. This is probably due to the fact that the diesel forklift was used to
420 load and unload trucks, which means that the forklift was moving from outdoor to
421 indoor having to drive by all the measurement points (worker area, indoor and outdoor).
422 Diesel and propane forklifts have been previously identified as a common source of
423 ultrafine particles (20 – 50 nm) in activities such as warehouse bagging and packing
424 (Ding et al., 2017; Huang et al., 2010; Kuhlbusch et al., 2004; Tsai et al., 2011; Viitanen
425 et al., 2017; Wang et al., 2010).

426 Finally, in terms of particle mass concentration, no increases when comparing to pre-
427 activity were detected for any of the four days as discussed before. However, during
428 the SB2 packing period, two peaks at 10:40 and 11:30 (Figure 4b) of particles at
429 around 1 μm which coincided with the start of an electric forklift were identified. Huang
430 et al. (2010) observed during packing of large bags (800 kg) that forklift activity
431 released considerable amounts of dust through particle resuspension. In the present
432 case, this phenomenon was only observed on one occasion, and therefore, no clear
433 relationship can be deduced between electric forklift activity and particle mass
434 concentration increases due to resuspension.

435

436

437 3.3. Exposure and risk assessment - Regional inhalation dose rates

438 Inhalation dose rates were estimated for each day using combined data from
439 NanoScan and MiniWras (Table 3 and S3). Particle number dose rates (\dot{n}) during
440 packing ranged between 682×10^6 and $1122 \times 10^6 \text{ min}^{-1}$. Increases (between 87×10^6 and
441 $240 \times 10^6 \text{ min}^{-1}$) during the packing process were obtained when comparing with pre-
442 activity periods for all days. Surface dose (\dot{s}) analysis was calculated as well as
443 respiratory tract deposition percentages. From the total surface area of the deposited
444 particles during packing ($3.3 - 7.6 \times 10^6 \text{ } \mu\text{m}^2 \text{ min}^{-1}$), 52 – 61% was estimated to deposit
445 in the alveolar region, 13 – 14% in the trachea bronchi and 25 – 36% in the head
446 airways (Table 3). The percentage for the alveolar region is lower than that found by
447 Wang et al. (2010), who determined the percentage of deposited surface area in the
448 alveolar region to be 80% during packing in a carbon black manufacturing industry. No
449 increases in the total surface deposited area during packing were observed when
450 compared with the pre-activity periods except for SB2. In addition, an increase on the
451 percentage on the alveolar and trachea bronchi regions during packing was observed
452 for SB1, whereas for the rest, percentages remained approximately the same. This
453 increase in number and surface deposited area is most likely due to the diesel forklift
454 activity or another process taking place near the packing area and not due to the
455 packing process itself, which emits coarser particles as described in previous sections.
456 The day with the highest percentage of diesel forklift activity (SB2) showed the highest
457 increase in total surface deposited area (4.6×10^6 and $6.0 \times 10^6 \text{ } \mu\text{m}^2 \text{ min}^{-1}$ for pre-activity
458 and process respectively). Higher percentages of deposited particles were detected in
459 the alveolar and head airways regions. Particles deposition on the tracheobronchial
460 area is dominated by particles with diameters under 10 nm. Here, instruments used
461 have an under limit at around 20 nm. Thus, when analyzing tracheobronchial
462 estimations the previous fact must be considered.

463 Particle number deposition percentages on the alveolar region ranged between 66 –
464 69%, similar range as in Wang et al. (2010), who found it to be 64% during packing in a
465 carbon black manufacturing industry. As pointed out in Wang et al. (2010) the use of
466 both metrics, number concentration and surface area, is advisable as, when used
467 separately, different results may be obtained. In Koivisto et al. (2012b) inhalation dose
468 rates as well as percentages of deposited particles in the respiratory tract were
469 calculated for nanoparticle production process in terms of particle number, mass and
470 active surface area. Increases in number concentration and surface area were
471 detected when comparing pre-activity period with process. For that specific case,
472 number concentration was found to be the metric defining better the particles emitted
473 during the process whereas surface area was found to describe process and
474 background particles (Koivisto et al., 2012b).

475 **3.4. Prediction models**

476 Exposure concentrations were modelled using the one and two box models including
477 and excluding outdoor concentrations. Worker area monitored concentrations were
478 compared to one box modelled results, and to FF modelled concentrations when using
479 the two box model, as worker area monitoring instruments were not placed inside the
480 limits of the defined NF area.

481 As described in section 2.4.3, a sensitivity analysis was carried out to identify the
482 optimal air flow rate between NF and FF (β) in the two box model for this industrial
483 setting. The range of values tested was obtained from the literature (Baldwin and
484 Maynard, 1998; Arnold et al., 2017), and the results of this analysis are summarized in
485 Tables 1 and 4. For small bags, a range of $S = 0.006\text{-}0.05 \text{ m s}^{-1}$, where S is wind
486 speed inside the plant, corresponding to $\beta = 0.36\text{-}3 \text{ m}^3 \text{ min}^{-1}$ was tested. Modelled
487 concentrations were seen to vary between 26 and 38%. On the other hand, for big
488 bags a range of $S = 0.0125\text{-}0.04 \text{ m s}^{-1}$ corresponding to $\beta = 9.4\text{-}30 \text{ m}^3 \text{ min}^{-1}$ was tested,

489 and modelled concentrations were seen to variate less than 5%. Results evidenced
490 that for small bags, higher β (e.g., $3 \text{ m}^3 \text{ min}^{-1}$) resulted in modelled/measured ratios up
491 to 1.89, whereas lower β largely underestimated modelled concentrations (ratios =
492 0.39-0.69 for $\beta = 0.36 \text{ m}^3 \text{ min}^{-1}$). As a result, a β of $0.75 \text{ m}^3 \text{ min}^{-1}$ was selected for the
493 small bag scenarios. In a similar analysis, for the big bag scenarios β was $30 \text{ m}^3 \text{ min}^{-1}$
494 (Table 1), although as explained β does not seem to be a critical parameter for this
495 scenario.

496 With the parametrization selected, for the one box setup including outdoor
497 concentrations, modelled concentrations ($325, 404, 759$ and $546 \text{ } \mu\text{g m}^{-3}$ for SB1, SB2,
498 BB1 and BB2, respectively) (Table 4) were able to reproduce actual exposure
499 measurements ($279, 318, 668$ and $528 \text{ } \mu\text{g m}^{-3}$ for SB1, SB2, BB1 and BB2,
500 respectively) (Table 3). Predicted concentrations were only slightly higher than the
501 measured values (Table 3 and 4). The ratio ($m_{modelled} / m_{measured}$) was 1.22 ± 0.07 for
502 the small bags and 1.09 ± 0.08 for big bags (Table 4). For the two box model including
503 outdoors, modelled concentrations ($311, 316, 745$ and $538 \text{ } \mu\text{g m}^{-3}$ for SB1, SB2, BB1
504 and BB2, respectively) (Table 4) were higher than measured concentrations with a ratio
505 ($m_{modelled} / m_{measured}$) of 1.05 ± 0.08 for small bags and 1.07 ± 0.07 for big bags
506 (Table 4).

507 Modelled concentrations without adding outdoor concentrations (Table 4) were
508 generally lower than measured concentrations (and only slightly higher in 2 cases; SB1
509 one and two box model including outdoor). The ratio ($m_{modelled} / m_{measured}$) for the one
510 box model was 0.98 ± 0.19 for the small bags and 0.84 ± 0.12 for the big bags. The
511 ratio ($m_{modelled} / m_{measured}$) for the two box model was of 0.88 ± 0.25 for the small
512 bags and 0.82 ± 0.12 for big bags. Thus, the model underestimated exposure
513 concentrations when outdoor contributions were not included. Commonly, model
514 testing assumes that the initial concentration is zero and that the supplied air is free of

515 contaminants (Zhang et al., 2009). However, as discussed in section 3.3.2, the
516 infiltration of outdoor contaminants is frequent, especially when having open doors as
517 in this case. In the industrial setting under study, modelled concentrations without
518 including outdoor were underestimated in 6 of the 8 cases. This kind of
519 underestimation has been considered detrimental in risk assessment (Arnold et al.,
520 2017). On the other hand, modelled concentrations when including outdoor slightly
521 overestimated measured concentrations and had higher precision. These more
522 conservative results were considered preferable from a risk assessment point of view.

523 Arnold et al. (2017) highlighted the importance of making the right model selection
524 when applying them to real cases. The use of the two box model in a well-mixed
525 environment can lead to an overestimation of the FF and especially of the NF modeled
526 concentrations, whereas using a one box model to estimate concentrations in a NF-FF
527 environment can lead to an underestimation. In the industrial setting under study, the
528 big bags scenario seemed to be clearly a one box case scenario due to the absence of
529 an enclosure. However, both models provided similar predictions, the one box model
530 resulting in only slightly higher concentrations. In general, overestimation by models
531 has been described for both, one and two box models (Johnson et al., 2011; Koponen
532 et al., 2015; Sahmel et al., 2009).

533 **4. Discussion**

534 Evidently, the results obtained regarding modelled concentrations are highly dependent
535 on model parameters such as the handling energy factor, local controls, air exchange
536 rate (AER) and NF-FF air flow (β), which are not yet fully parametrized (Cherrie et al.,
537 2011; Jayjock et al., 2011; Sahmel et al., 2009; Baldwin and Maynard, 1998; Keil and
538 Zhao, 2017) and are often challenging to estimate (Zhang et al., 2009). Sensitivity
539 analyses such as the one presented in Table 1 are also valuable.

540 In the case of the AER and β , experimental data were not available for this case study
541 and they were thus obtained from the literature and tested by means of a sensitivity
542 analysis. β was seen to be a key parameter when modelling the small bags case
543 scenario while it is not critical for the big bags case scenario. That may be explained by
544 the fact that the small bags case scenario was a real two box case (with an actual
545 enclosure and with a small surface area for the air flow between NF and FF) whereas
546 for the big bags there was no real separation between NF and FF and consequently
547 the theoretical free surface area used in the model was much higher.

548 Local controls prevent dispersion of the aerosolized particles in the room air or remove
549 the particles from air, e.g. enclosures or local extraction systems (Fransman et al.,
550 2008). When having to consider extractions systems, local control values associated
551 can be relatively easy to determine, but in cases like enclosures or barriers it is more
552 complex especially without having actual exposure concentrations. Local exhaust
553 ventilation efficiency can be calculated by a relatively simple equation (Hewett and
554 Ganser, 2017) although some unknown parameters are required. For cases such as
555 the present study when there is no possibility to experimentally establish a value,
556 Fransman et al. (2008) conducted a review with values proposed for different local
557 controls. Here, enclosure local control and bag protection was included in the equation
558 by using values reviewed in Fransman et al. (2008). The output modelled
559 concentrations were seen to correctly predict measured concentrations when using the
560 reported values.

561 Finally, the emission source characterization is one of the main sources of uncertainty
562 in the model, as it is strongly case-specific. This is one of the reasons why studies
563 dealing with real-world scenarios are highly necessary in the literature. As in the
564 present study, emission source characterization can be based on the dustiness index
565 which may be obtained by standard methods (Lidén, 2006). However, the handling
566 energy factor must be considered (Koivisto et al., 2015; Lidén, 2006; Schneider and

567 Jensen, 2007). When the dustiness concept cannot be used, equations to estimate
568 emission rates have been described (Hewett and Ganser, 2017; Sachse et al., 2012)
569 and used on real scenarios by using mass equation balance and a convolution theorem
570 (Koivisto et al., 2018a; Koivisto et al., 2018b). However, unlike the other parameters,
571 literature regarding emission rates is still limited.

572 Additionally, an important consideration to be discussed at this point is that the models
573 do not consider particle losses due to sedimentation. Cherrie et al. (2011) found that for
574 particles $< 10 \mu\text{m}$ the impact of deposition might be reasonably ignored, but for
575 particles with a higher aerodynamic diameter the deposition impact may be important.
576 Figure 5 shows that most of the emitted particles during packing were under $10 \mu\text{m}$.
577 However, for BB1, a slight increase of particles $> 10 \mu\text{m}$ during packing was observed.

578 Based on the considerations above, it may be concluded that the use of the one box
579 and two box models in the industrial setting tested can satisfactorily predict particle
580 concentrations, especially when input parameters are sufficiently robust. In Sahmel et
581 al. (2009), the steady state model, similar to the one box model used here, was seen to
582 correctly perform concentration modeling when choosing the appropriate factors.
583 However, in industrial settings many considerations must be taken into account and
584 what is clearly observed in a laboratory scale or controlled settings cannot be directly
585 extrapolated to the industrial world. To this end, the parameters used in this work and
586 the coefficients applied, described in section 2.4, may be useful as input for future
587 modelling studies.

588 **5. Conclusions**

589 Packing of a fertilizer into small (respirable fraction range $279\text{-}318 \mu\text{g m}^{-3}$) and big bags
590 (respirable fraction range $487\text{-}668 \mu\text{g m}^{-3}$) was not seen to significantly increase worker
591 exposure compared with pre-activity concentrations in terms of inhalable and respirable
592 concentrations. However, increases in particle number concentrations were observed,

593 quite likely related to the diesel forklift activity. A statistically significant increase in
594 ultrafine particles was observed for SB2 (58646 cm⁻³ during pre-activity; 75912 cm⁻³
595 during packing). This dataset was used to test the performance of one and two box
596 models as tools for risk assessment under real-world industrial settings.

597 The one and two box models were tested in a real industrial exposure case scenario,
598 during packing of a fertilizer into small and big bags, with and without enclosure. Both
599 models seemed to be able to predict exposure concentrations. When outdoor
600 concentrations were not included in the models, modelled concentrations slightly
601 underestimated actual concentrations, with ratios modelled/measured ranging between
602 0.82 ± 0.12 and 0.98 ± 0.19 for the respirable size fraction. The use of outdoor
603 concentrations as an input for the models was seen to improve model performance,
604 resulting in slight overestimations of measured concentrations what was estimated as
605 preferable from a risk assessment point of view. In addition, higher precision between
606 repetitions was achieved when including outdoor contributions (ratio
607 modelled/measured 1.05 ± 0.08 to 1.22 ± 0.07). Thus, it was concluded that including
608 outdoor concentrations in the model resulted in an improved model performance, which
609 may be considered a step forward in the application of risk assessment models.

610 With regard to the selection of the one or two box models, similar results for the small
611 and big bags case scenarios were obtained. However, slightly better results were
612 obtained when using the two box model for the small bags scenario (one box model
613 1.22 ± 0.07 ; two box model 1.05 ± 0.08), whereas both models provided similar results
614 for the big bags (1.09 ± 0.08 and 1.07 ± 0.07 respectively). Thus, it may be concluded
615 that, even in complex real-world settings, the simplest approach of the one box model
616 may provide good results if it is adequately parametrized. Model parametrization is in
617 itself a key issue: the selection of parameters such as the handling energy factor, the
618 local controls and especially the NF-FF air flow in the two box model were seen to be
619 critical for the model's performance. Here, NF-FF air flow, local controls efficiency as

620 well as handling energy factor were assumed based on literature databases, and
621 relatively accurate predictions were obtained. Therefore, reporting measured or tested
622 values for these parameters is seen as necessary to expand the use and applicability
623 of prediction models for risk assessment.

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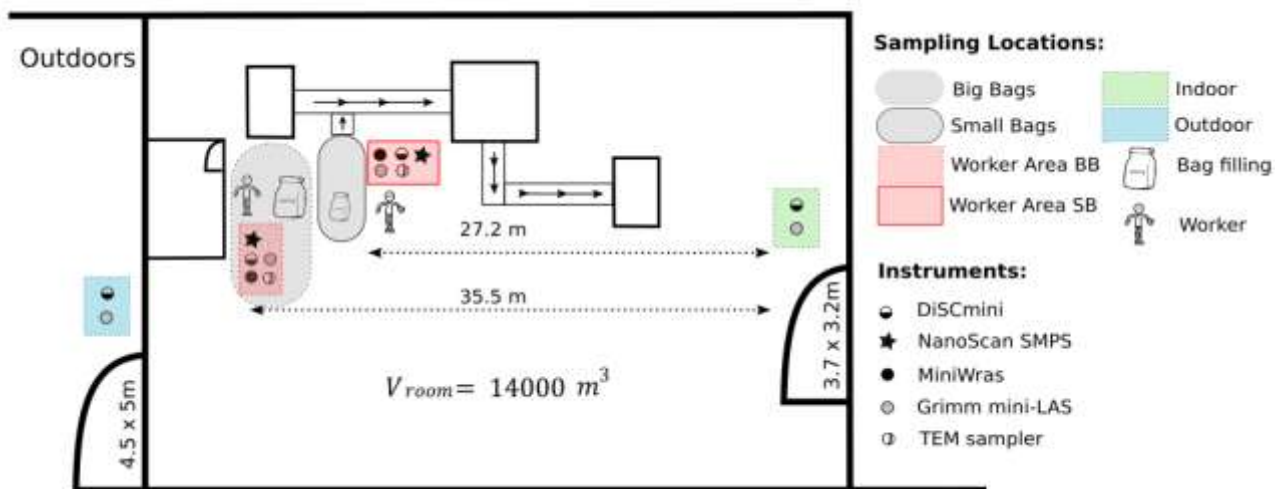
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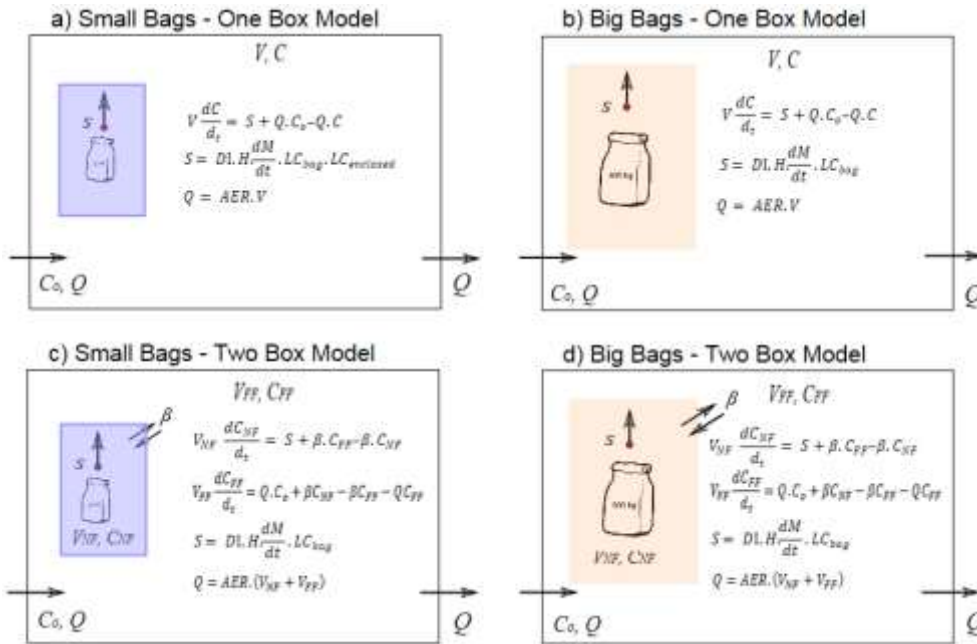
828 **Figures and Tables**



829

830 **Figure 1.** Packing industrial unit layout. Measurement locations as well as devices used during packing
 831 operation are pointed out. BB: big bags. SB: small bags.

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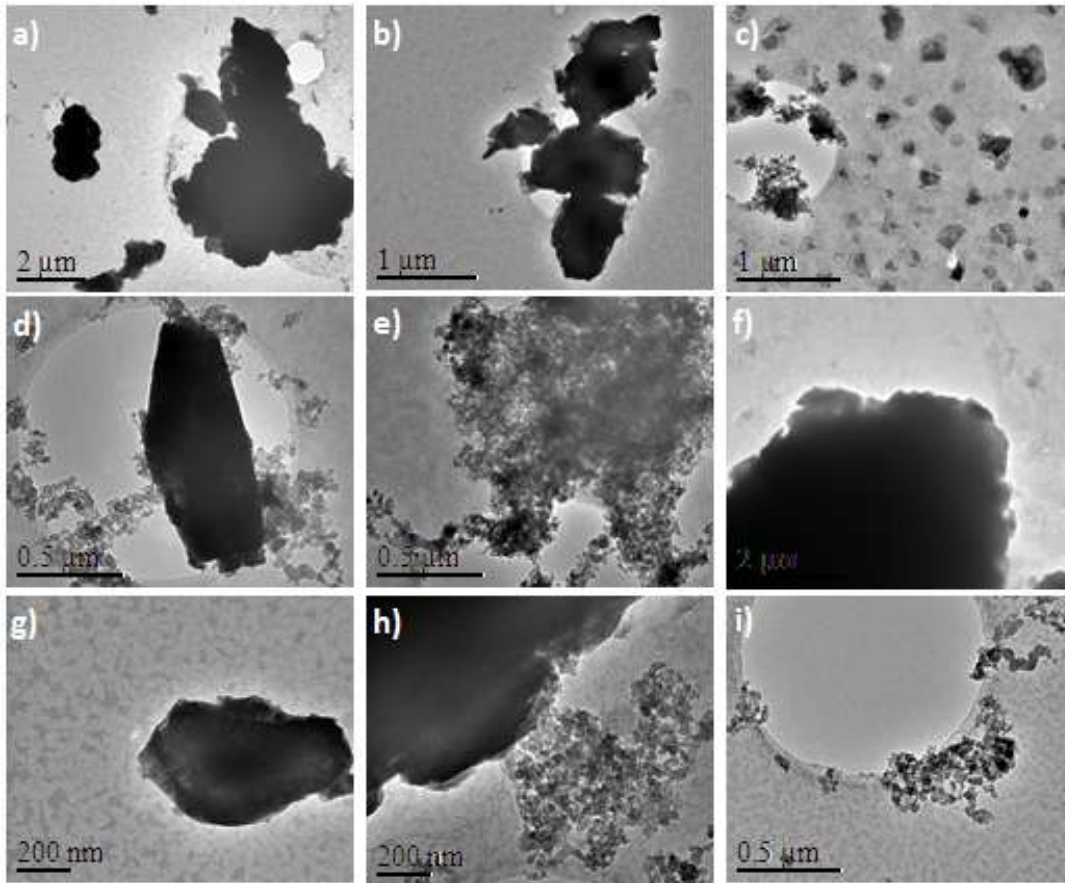


SMALL BAGS			
Respirable dustiness index	DI	16 mg/kg	Very low dustiness index
Mass flow	dM/dt	250 kg/min	Mass flow value
Handling energy	H	0.5	Drop height is approximately half of the dustiness test drop height
Local emission controls	LC _{bag}	0.6	0.6 = bag acts as a protection (reduction of the emissions of a 40%)
	LC _{enclosed}	0.5	0.5 = process enclosed (reduction of the emissions of a 50%)
Room volume	V _{room} /V _{FF}	14000 m ³	Big industrial unit
Air Exchange rate	AER	7 ACH	Medium ventilation rate
Near field volume	V _{NF}	6 m ³	Volume of the enclosed space, it does not include the worker area
Near field air flow	β	0.75 m ³ /min	Low air exchange NF-FF (enclosed process)
BIG BAGS			
Respirable dustiness index	DI	16 mg/kg	Very low dustiness index
Mass flow	dM/dt	175 kg/min	Mass flow value
Handling energy	H	1	1 is equivalent to the drop height in the dustiness test
Local emission controls	LC _{bag}	0.6	0.6 = bag acts as a protection (reduction of the emissions of a 40%)
Room volume	V _{room} /V _{FF}	14000 m ³	Big industrial unit
Air Exchange rate	AER	7 ACH	Medium ventilation rate
Near field volume	V _{NF}	25m ³	Volume of the bagging line area including worker area
Near field air flow	β	30 m ³ /min	High air exchange NF-FF (opened process)

833 **Figure 2.** One box and two box model description and parameters values (table). For small bags, one box

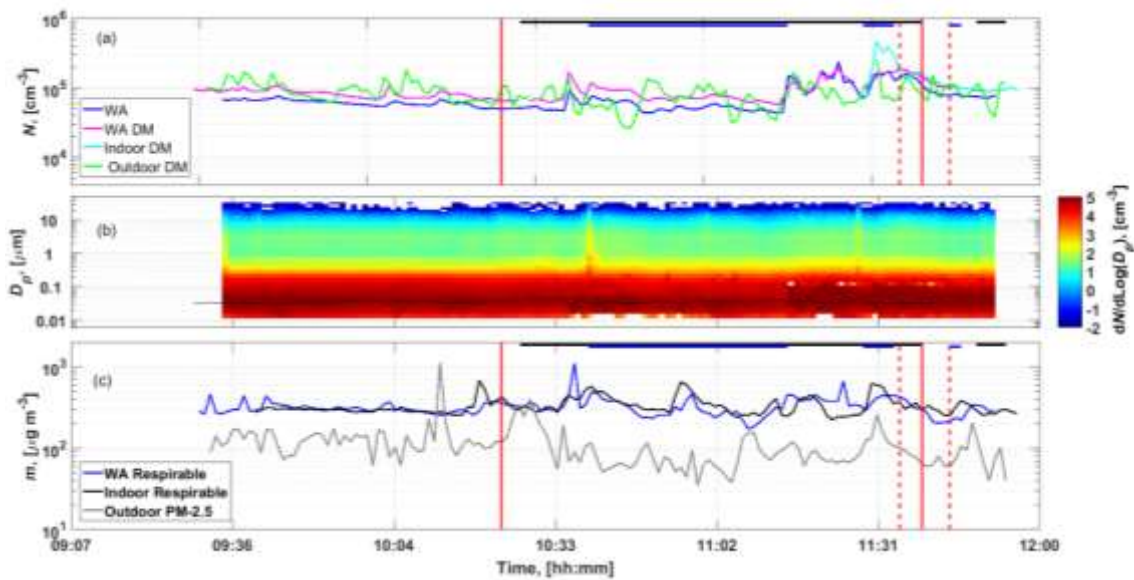
834 model (a) and two box model (c) and for big bags, one box model (b) and two box model (d).

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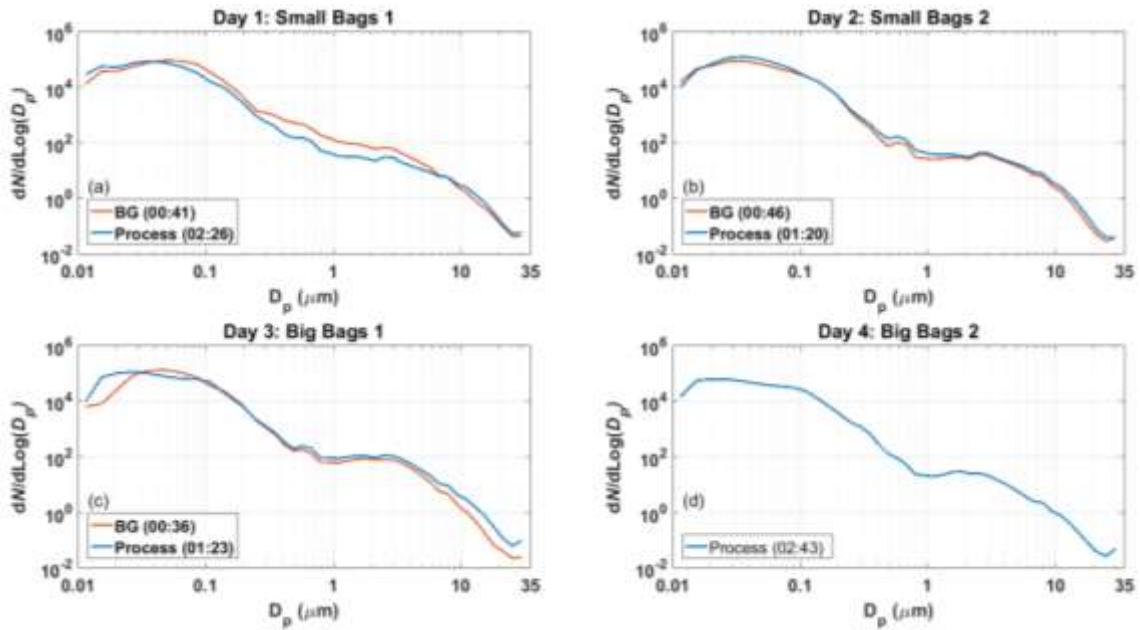
837 **Figure 3.** TEM images of the collected particles during the fertilizer bag filling process with small (a, b, c, d
 838 and e), and big bags (f, g, h and i).



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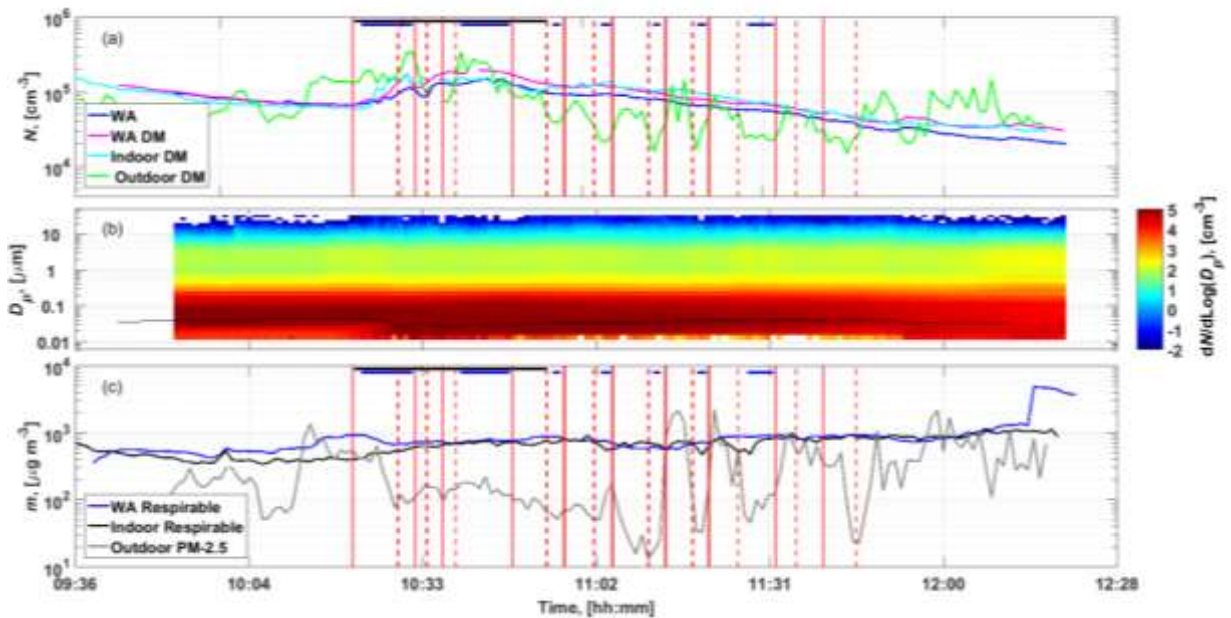
840 **Figure 4.** Particle concentration at packing area (WA) during small bags 2 (SB2): (a) particle number
 841 concentration time series; (b) particle size distribution time series measured with the MiniWras and the
 842 NanoScan, solid black line shows DiSCmini (DM) D_{50} ; (c) mass concentration time series. Red vertical

843 lines indicate start (solid line) and stop (dashed line) of the packing operation and horizontal black and
 844 blue lines in the top of the graphs indicate diesel and electric forklifts activity respectively.



845

846 **Figure 5.** Average particle size distributions measured by NanoScan and MiniWras during pre-activity and
 847 packing processes for (a) small bags day 1, SB1; (b) small bags day 2, SB2; (c) big bags day 1, BB1 and
 848 (d) big bags day 2, BB2.



849

850 **Figure 6.** Particle concentration at packing area (WA) during big bags day 1 (BB1): (a) particle number
 851 concentration time series; (b) particle size distribution time series measured with the MiniWras and the
 852 NanoScan, solid black line shows DiSCmini (DM) D_{50} ; (c) mass concentration time series. Red vertical

853 lines indicate start (solid line) and stop (dashed line) of the packing operation and horizontal black and
 854 blue lines in the top of the graphs indicate diesel and electric forklifts activity respectively.

855 **Table 1.** Sensitivity analysis for different air flow values for small and big bags with ratios (modelled
 856 values/measured values). Variation (%) of the modelled concentration when using the higher and lower air
 857 flow value is reported. $\beta = \frac{1}{2} \cdot SA \cdot S$; where *SA* is the surface area.

β (m ³ min ⁻¹)	S (m s ⁻¹)	SB1		SB2		β (m ³ min ⁻¹)	S (m s ⁻¹)	BB1		BB2	
		With outdoor Ratio	Ratio	With outdoor Ratio	Ratio			With outdoor Ratio	Ratio	With outdoor Ratio	Ratio
0.36	0.006	0.69	0.63	0.68	0.39	-	-	-	-	-	-
0.5	0.004	0.87	0.81	0.81	0.51	-	-	-	-	-	-
0.75	0.0125	1.11	1.06	0.99	0.70	9.4	0.0125	1.08	0.70	0.99	0.88
1	0.017	1.30	1.25	1.15	0.86	12.75	0.017	1.10	0.71	1.00	0.89
1.5	0.025	1.56	1.50	1.40	1.1	15	0.02	1.10	0.71	1.01	0.90
1.8	0.03	1.66	1.60	1.51	1.21	-	-	-	-	-	-
2.4	0.04	1.80	1.75	1.67	1.36	30	0.04	1.12	0.73	1.02	0.91
3	0.05	1.89	1.84	1.79	1.50	-	-	-	-	-	-
Variation (%)		36.4	34.6	38.0	25.8	Variation (%)		3.0	4.5	2.6	2.9

858

859 **Table 2.** Pre-activity, total process time, and total time for each activity (packing, electric forklift and diesel
 860 forklift) shown in hh:mm. The percentage of time of each activity (packing, electric forklift and diesel forklift)
 861 with respect to the total length of the process is included in brackets. Background period (pre-activity) not
 862 included. Less than 5 minutes difference between stop and start of the next activity was counted as the
 863 same period activity.

Process	Background time	Packing process time	Material pouring time (%)	Electric forklift time (%)	Diesel forklift time (%)
SB1	00:41	02:26	02:03 (84.2%)	00:02 (1.4%)	00:44 (30.2%)
SB2	00:46	01:20	01:16 (95.0%)	00:42 (53.2%)	01:17 (96.2%)
BB1	00:36	01:23	00:46 (55.9%)	00:26 (32.1%)	00:31 (38.4%)
BB2	-	02:43	00:36 (22.1%)	00:40 (24.9%)	02:05 (77.1%)

864 **Table 3.** Mean number concentration and mass concentrations during background period (BG) (pre-
865 activity) and packing process in the worker area (WA). N_{TOT} (D_p : 10 nm – 35 μ m), N_{UPF} (D_p < 86.60 nm),
866 N_{FP} (86.60 nm < D_p < 943.0 nm), N_C (D_p > 943.0 nm). Mass concentrations are shown in terms of
867 inhalable, thoracic and respirable fractions measured with the Grimm monitor. Calculated dose rates in
868 particle number, \dot{n} , and surface area, \dot{s} , and regional deposition in percentages on head airways, trachea
869 bronchi and alveolar. Values in bold indicate statistically significant differences compared with background
870 concentrations.

	Small Bags day 1		Small Bags day 2		Big Bags day 1		Big Bags day 2	
	(SB1)		(SB2)		(BB1)		(BB2)	
	BG	Packing	BG	Packing	BG	Packing	BG	Packing
N_{TOT} [cm^{-3}]	67254 \pm	63108 \pm	63797 \pm	81137 \pm	81073 \pm	85949 \pm	-	50290 \pm
	11076	29592	5435	42448	8719	29748	-	40893
N_{UPF} [cm^{-3}]	61083	59900	58646	75912	73641	77945	-	46359
N_{FP} [cm^{-3}]	6121	3188	5129	5197	7383	7935	-	3922
N_C [cm^{-3}]	50	20	22	28	50	68	-	14
Inhalable [$\mu\text{g m}^{-3}$]	1987 \pm	2025 \pm	1866 \pm	1276 \pm	1650 \pm	1864 \pm	-	1047 \pm
	214	975	1141	550	588	556	-	923
Thoracic [$\mu\text{g m}^{-3}$]	1487 \pm	1053 \pm	1147 \pm	962 \pm	1183 \pm	1507 \pm	-	787 \pm
	138	435	315	345	367	381	-	721
Respirable [$\mu\text{g m}^{-3}$]	634 \pm	279 \pm	362 \pm	318 \pm	354 \pm	668 \pm	-	528 \pm
	64	131	74	109	105	153	-	898
$\dot{n}_{, \cdot 10^6}$ [min^{-1}]	770	857	834	1035	882	1122	-	682
$\dot{s}_{, \cdot 10^6}$ [$\mu\text{m}^2 \text{min}^{-1}$]	6.4	4.3	4.6	6.0	7.5	7.6	-	3.3
$\dot{s}_{, \cdot}$ Head airways [%]	30.7	26.3	27.1	26.2	28.0	36.0	-	24.7
$\dot{s}_{, \cdot}$ Trachea bronchi [%]	12.7	14.2	13.9	14.0	13.3	12.5	-	14.2
$\dot{s}_{, \cdot}$ Alveolar [%]	56.6	59.5	59.0	59.8	58.7	51.5	-	61.1

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875 **Table 4.** One box and two box modeled respirable concentration results including and without including
876 outdoor concentrations. Ratios between modeled and measured concentrations for each specific case are
877 shown in brackets. Last two columns are mean ratio values (and standard deviation) for small and big
878 bags (SB and BB). Last row shows the measured respirable fraction concentrations in the workers area
879 and the spatial background.

[$\mu\text{g}/\text{m}^3$] (ratio modelled/measured)	SB1	SB2	BB1	BB2	Ratio mean \pm (s.d)	
					Small bags	Big bags
One box with outdoor	325 (1.16)	404 (1.27)	759 (1.14)	546 (1.03)	1.22 (0.07)	1.09 (0.08)
Two box with outdoor (FF)	311 (1.11)	316 (0.99)	745 (1.12)	538 (1.02)	1.05 (0.08)	1.07 (0.07)
One box	310 (1.11)	270 (0.85)	501 (0.75)	488 (0.92)	0.98 (0.19)	0.84 (0.12)
Two box (FF)	296 (1.06)	223 (0.70)	487 (0.73)	480 (0.90)	0.88 (0.25)	0.82 (0.12)
Measured respirable fraction in Worker Area	279	318	668	528	-	

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