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1 **Evaluation of particulate matter (PM₁₀) emissions and its chemical characteristics during**
2 **rotary harrowing operations at different forward speeds and levelling bar heights**

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8

9 **Abstract**

10 Particulate matter (PM) is an air pollutant which poses a considerable risk to human health. The
11 agricultural sector is responsible of the 15% of the total anthropogenic emissions of PM₁₀ (PM
12 fraction with aerodynamic diameter below 10 µm) and soil preparation activities have been
13 recognized as one of the main drivers of this contribution. The emission factors (EF) proposed
14 by European environmental agency (EEA) for tilling operations are based on very few studies,
15 none of which has been made in Italy. Moreover, few studies have considered the influence of
16 operative parameters on PM₁₀ emissions during tilling. The aim of this work was to assess PM₁₀
17 emission and dispersion during rotary harrowing and to understand how operative parameters,
18 such as forward speed and implement choice may affect PM release. A further objective was
19 to assess the near field dispersion of PM₁₀ to address exposure risks. Emission factors (EFs)
20 were determined during two different trials (T1 and T2). During T1, the effect of tractor speeds
21 (0.6, 1.1 and 1.7 m s⁻¹) on PM₁₀ emissions was investigated, while in T2 a comparative essay
22 was made to study the influence of levelling bar height on emissions. The average ground level
23 downwind concentrations of PM₁₀ during harrowing operation was estimated through dispersion
24 modelling. The observed PM₁₀ EFs for rotary harrowing were 8.9 ± 2.0 mg m⁻² and 9.5 ± 2.5
25 mg m⁻² on T1 and T2, respectively. The heavy metal content of soil-generated PM₁₀ was also
26 assessed. In the generated PM, the elemental concentrations were higher than ones in soil.
27 As, Cd and Ni concentration levels, determined in PM₁₀ near to the tractor path, were also high,

28 being several times higher than the annual average regulatory threshold levels in ambient air,
29 as defined by the European regulation.

30

31 KEYWORDS: PM₁₀; emission factors; metals; soil preparation; harrowing

32

33 CAPSULE: PM₁₀ emission factors for rotary harrowing have been determined with different
34 tractor speeds, and relevant metal contents were found in emitted PM₁₀.

35

36 **1. Introduction**

37 The increase in atmospheric concentrations of particulate matter (PM) is a major cause of
38 concern, having been associated with acute and chronic health effects and even with the rise
39 in mortality and morbidity rates (WHO, 2006; Tonne et al., 2016). Many emission sources
40 contribute to PM₁₀ (PM fraction with aerodynamic diameter below 10 µm), among which
41 agriculture may play a significant role, being held responsible for the 15% of total anthropogenic
42 PM₁₀ emissions in Europe (EEA, 2019). Agricultural emissions of primary particulate mainly
43 derive from wind erosion of agricultural soils, livestock farming activities and crop management
44 (Maffia et al., 2020). Crop management activities have been recognized to be a substantial
45 contributor to the overall emissions (Sharratt and Auvermann, 2014). Currently, the emission
46 factors (EF) used by the European Environmental Agency (EEA) for crop management
47 operations are based on a limited number of studies and did not take into account the different
48 tilling implements used by farmers. Moreover, few studies have considered the influence of
49 operative parameters on PM₁₀ emissions during tilling (Maffia et al., 2020).

50 Issues related to PM emissions and atmospheric concentrations have recently been at the
51 centre of public attention in Northern Italy due to the associated health risks. In fact, the latest
52 report of the Italian institute for environment protection and research has highlighted
53 exceedances of the recommended daily PM₁₀ concentration threshold (50 µg m⁻³) for more than
54 35 days per year in most monitoring stations of the Po Valley (Cattani et al., 2019).

55 The northwest part of this area, where the present trials took place, is characterized by a low
56 average annual wind speeds, which fall often below 1 m s⁻¹ (Fratianni et al. 2007), and by being

57 intensively exploited, both by industrial and farming activities, and densely populated. To face
58 the high PM pollution of the area (Cattani et al., 2019) it is important to acknowledge its specific
59 climatic conditions and to start assessing local emission factors for the main emission sources,
60 to provide the policy makers with up to date information to address the air quality issue.

61 Health risks linked to PM are not only due to the size of the particles or to the concentration,
62 but also to its elemental composition (Kendall et al., 2004). Particularly, many studies have
63 focused on the potentially toxic effects due to Trace Elements (TE) adsorbed on PM₁₀ in urban
64 and roadside environments (Padoan et al., 2016; Zhang et al., 2019; Wu et al., 2020).

65 Agricultural soils are well known for being both sources of PM₁₀ and, at least in certain areas,
66 enriched in TE due to both anthropogenic and natural sources (Li et al., 2019). In fact, the
67 application of pest control products and organic fertilizers, such as pig manure, has been shown
68 to increase the soil reserves of TE such as Cu, Zn and Mn (Brun et al., 2001; Guo et al., 2018).
69 Nevertheless, few information is available on the concentrations of these elements in the
70 airborne PM₁₀ emitted during tilling or wind erosion events. A recent investigation (Wang et al.,
71 2016) has shown that the TE concentrations in PM₁₀ of 4 different agricultural regions in China
72 were higher than the expected, with carcinogenic risk above the acceptable limits due mostly
73 to Pb, Co, Ni and Cd concentrations. It is therefore important to consider particle composition
74 when assessing PM emissions from agricultural sources.

75 The main aim of this study was to improve the knowledge on PM₁₀ emissions during soil
76 preparation operations and, in particular, on those due to rotary harrowing. Emissions from
77 rotary harrowing (coupled with packer roller and with levelling bar) were assessed in low wind
78 conditions, to provide a local EF for this operation, which has been poorly studied before.
79 Further objectives were to assess the effect of operative parameters, such as tractor speed and
80 levelling settings on the emission value. In addition, the characteristics of the emitted PM₁₀ and
81 their near field dispersion were assessed to obtain a broader picture of the impact that
82 harrowing operations can have on human health.

83 The field experiments presented hereafter are the first assessments of PM emission from land
84 preparation activities performed in Northern Italy. In this specific area, the environmental,
85 topographic, and demographic conditions could heavily influence both the amount of emission
86 related to soil cultivation and their potential contribution to the total PM exposure levels.

87

88 **2. Materials and methods**

89 *2.1. Experimental layout and Field measurements*

90 Two different trials, T1 and T2, were performed in July and October 2019, respectively, in two
91 different locations of the Piemonte region, Italy (44°50'27.9" N, 7°21'32.2" for T1;
92 44°54'52.9" N, 7° 23' 45.9" E for T2) in two fields with a sandy-loam soil for T1 trials and a
93 loamy soil for T2 trials. In both trials, measurements of PM₁₀ were carried out at each tractor
94 passage using an optical PM monitor (TSI, DustTrack™ II model 8530), with a sampling
95 frequency of 1 Hz. The PM monitor was placed alongside the area tilled by the tractor at 4 m
96 (Figure 1). The instrument was moved to the next passage line after each pass and placed
97 either east or west of the line according to wind direction. The DustTrack monitor was placed
98 in the field 1 hour before the start of the trial and continued sampling until 1 hour after the trial,
99 to assess the background PM₁₀ concentration.

100 The positioning of the instrument was arranged similarly to what done in previous studies
101 (Faulkner, 2013; Kasumba et al., 2011), with the sampler inlet placed at 1 m aboveground and
102 with a fixed distance between the sampler and the tractor path of 4 m. According to the results
103 of Holmén et al. (2008) and Kasumba et al. (2011), obtained in New Mexico, sampling at higher
104 distances from the source could lead to underestimate the concentration of finer PM fractions
105 due to vertical dispersion of the plume and to the increased distance between the sampler inlet
106 and the plume centre.

107 Meteorological data were collected using a weather station mounted in a corner area of the
108 field, with every side free from obstacles. The weather station has two 3D anemometers
109 (Campbell scientific, 3D Metek uSonic-Omni), mounted at 2 and 4 m above ground
110 respectively, and a temperature probe (HOBO, U12). The anemometers data were sampled at
111 a rate of 5 Hz.

112 Field trials were carried out with a 12 rotors, 3 m working width, rotary power harrow (Frudent
113 Eternum R303-19, Frudent Group s.r.l., Italy). The harrow was equipped with a packer roller
114 (0.55 m diameter) on T1, whereas a levelling bar was installed in T2 in order to evaluate EF in
115 different implement configurations. In T2, the roller was replaced with a couple of wheels
116 mounted on the same tillage depth adjustment system.

117 The rotary harrow was hooked up to the three point hitch of a four-wheel-drive row crop tractor
118 (Fendt 718 Vario, AGCO GmbH, Germany) having a 132 kW maximum engine power and an
119 unladen mass of 7155 kg (OECD, 2010). A ballast of 1200 kg was also linked to the front three
120 point hitch in order to reduce wheels slip.

121 During harrowing PTO rotation speed was maintained at about 1000 rpm achieving a rotor
122 angular speed of 285 rpm, while the tillage depth was set to 10 cm.

123 In T1, 36 passages were performed, although some of those had to be later excluded from the
124 analyses due to sudden changes in wind direction that resulted in imprecise EF estimations
125 (the final number of calculated passages was 32). The length of each harrower passage has
126 been, in both cases, of 40 m. The experimental layout was designed in order to test the effect
127 of three different tractor speeds ($S1 = 0.6 \text{ m s}^{-1}$, $S2 = 1.1 \text{ m s}^{-1}$ and $S3 = 1.7 \text{ m s}^{-1}$), where S2
128 is the one normally implied by farmers, on PM_{10} emissions. S1, S2 and S3 passages were
129 randomized inside large plots (3 m wide and 120 m long), that were considered as blocks for
130 the statistical analysis and served to the purpose of limiting the variability linked with soil
131 heterogeneity and wind speed. The scheme of a large plot layout and of PM sensor positioning
132 is represented in Figure 1.

133 In T2, 24 tractor passages were performed (two of those were lost due to sudden wind direction
134 changes). The experimental layout was designed in order to test the effect of levelling bar height
135 on PM_{10} emissions. The bar attachment height was alternatively adjusted to two different levels,
136 a lower, L, and a higher one, H. The attachment height of the bar was tested both with the bar
137 perpendicular to the ground (S) and inclined of a 45° angle (I). In addition, the distance of the
138 bar from the harrower was varied during the trial among D1 (23 cm from the harrower) and D2
139 (28 cm from the harrower). The combinations of bar attachments heights and orientations
140 resulted in different distances between the bar and the point of the harrower teeth, being of 14,
141 16.5, 17 and 20 cm for L-S, L-I, H-S and H-I respectively. The different configurations and the
142 split plot experimental layout are graphically represented in Figure 2. The plot was organized
143 so as to reduce the time-lapse among passages involved in direct comparison, limiting the
144 variability linked with changing environmental conditions (such as wind speed). The levelling
145 bar is commonly installed on rotary harrows to improve soil fragmentation during harrowing by
146 keeping the soil closer to the harrow rotors for a longer period. The variation of the vertical
147 position and of the distance of the bar from rotors will change soil interaction with rotors tines,

148 modifying aggregates size and affecting PM emissions (Madden et al., 2010). Bar inclination,
149 instead, was an experimental solution aimed to reduce draught and, therefore, fuel
150 consumption.

151 Soil conditions in T1 and T2 were different. In T1 harrowing was performed on bare soil after
152 tillage, while on T2 only a superficial incorporation of crop residues (maize stalks) had been
153 performed, leaving a rougher surface with some residues still on the surface. In both cases soil
154 samples were taken at 0-15 cm depth.

155 At each sampling site, sub-samples were collected in the center of each parcel (36 sub-samples
156 in T1 and 24 in T2), mixed into one sample and quartered in field. Soils were dried at room
157 temperature and sieved with a 2-mm sieve prior to laboratory analyses.

158

159 2.2. *Elaboration of meteorological data*

160 Start and end times of each tractor passage were recorded during field measurements. The
161 passage time intervals were used to clip the output file of the anemometer, to obtain the average
162 wind components (u , v , w) at the time of each peak. Wind components were then used to
163 assess main wind speed (WS) and wind direction (WD) according to Stull (2012).

164 The Pasquill Guifford class (PG_{class}) was estimated for each passage by first calculating the
165 Monin Obhukov Length (L_{length}) and then estimating the stability class according to the table in
166 Smith et al. (1995). The L_{length} was estimated using the Bigleaf R package (Knauer et al., 2018),
167 according to the method described in Foken (2008). The input parameters required by Bigleaf
168 were air temperature (T_{air}), atmospheric pressure (p), friction velocity (u^*) and sensible heat flux
169 (H). T_{air} was retrieved by field measurements; p was assessed from elevation using Bigleaf
170 package (Knauer et al., 2018); u^* was calculated from wind data according to the method in
171 Stull (2012); H was assessed on the base of the estimation procedure proposed by Hanna and
172 Chang (1992).

173

174 2.3. *Dispersion modelling to estimate EFs and downwind concentration increases*

175 EFs were estimated using a backward lagrangian model (WindTrax). The input parameters to
176 the model were WS (at 2 and 4 m of height), WD (at 2 and 4 m of height), PG_{class} , T_{air} , average
177 PM_{10} concentration (at four meters from the source) and average background PM_{10}
178 concentration. The model was set to simulate the dispersion of 10^6 particles and the surface
179 roughness (Z_0) was set to the reference value (1 cm), parameterized for “bare soil” conditions.
180 The emission source was modelled as an area source having the same dimension of the plot
181 tilled at each harrower passage. Modelling tilling sources as areas rather than moving point
182 sources is the most common solution for EFs estimation through backward modelling (Faulkner,
183 2013; Funk et al., 2008; Jahne et al., 2015).

184 A simulation was ran per each tractor passage performed on the two field days. The output
185 given by the model was an Emission Rate (ER, $mg\ m^{-2}\ s^{-1}$) referred to the modelled area
186 source. The ER was later converted in EF ($mg\ m^{-2}$) according to the following formula:

$$187 \quad EF = ER\ t_{pass}$$

188 Where t_{pass} is the elapsed time (s) between the start and the end of each passage. The above
189 equation follows the principle presented by Faulkner et al. (2009).

190 Near source concentration increases during harrowing have been estimated and plotted using
191 the GRAL model (using its open source graphical user interface, GUI), which is a high resolution
192 lagrangian model and has previously been used to assess PM dispersion from tilling (Funk et
193 al., 2008). Moreover, the model has been proved to be particularly suited for modelling under
194 low wind speed conditions (Öttl et al., 2005; Öttl et al., 2002). Two dispersion simulations (one
195 for T1 and one for T2) were realised considering for both an equal area source, with a surface
196 of 1 ha and a squared shape, so that the concentration increases can be related to a known
197 area source of regular size and properly compared. The main inputs to the model were WS,
198 WD, PG_{class} and the PM_{10} emission rate ($kg\ h^{-1}$). The average WS and the prevalent WD
199 observed during D1 and D2 passages were used to run two different simulations. The stability
200 classes used were B and D for T1 and T2, respectively. The ER was obtain converting the
201 estimated EFs on both days into $kg\ ha^{-1}$ and considering that harrowing a surface of 1 ha at an
202 average speed of $3\ km\ h^{-1}$ requires 1 h of time. Downwind concentration was estimated at a
203 height of 1 m (over the ground level) and the horizontal grid resolution was of 1 m.

204 2.4. Soil analysis

205 All samples were analysed for pH (1:2.5 soil:water), total carbon and nitrogen (TC, TN)
206 (UNICUBE, Elementar), carbonates (volumetric method), bulk density and field humidity
207 according to the official Italian methods (Colombo & Miano, 2015). The particle-size distribution
208 (PSD) was measured via the sieve-pipette method (Gee and Bauder, 1986).

209 To determine the pseudo-total metal content in soil, *aqua regia* (HCl:HNO₃ 3:1 v/v) microwave
210 extraction was performed (Ethos D, Milestone). The elemental pseudo-total contents of 22
211 elements (listed in Table 3) were determined in all samples using ICP-MS (NexION 350D,
212 Perkin Elmer). All analyses were performed in duplicate. Accuracy was verified using a Certified
213 Reference Material for *aqua regia* soluble contents (CRM 141R, Community Bureau of
214 Reference, Geel, Belgium). Recoveries were between 95 – 105 % for all elements. All reagents
215 were of ultrapure or analytical grade.

216

217 2.5. Soil resuspension in laboratory and PM₁₀ filter analyses

218 The soils collected in the field, after being dried as for chemical analyses, were re-suspended
219 under laboratory condition to simulate PM₁₀ emission during tilling. Soil was re-suspended using
220 a soil resuspension chamber, which was assembled using a rotating plastic (PET) drum
221 (Madden et al., 2010), having a cylindrical form, a total volume of 25 L and a circular opening
222 of 15 cm of diameter. The drum was moved by an electric engine (0.75 kW) at an average
223 speed of 26 rpm. A filter based high volume sampler (TCR Tecora[®], Echo Hi-Vol), working with
224 a 220 L min⁻¹ flow rate, was placed in front of the drum opening to sample the out coming PM₁₀
225 particles. The scheme of the resuspension system is illustrated in Figure 3.

226 A sample of 1 kg of soil was re-suspended for each trial (T1 and T2) and the re-suspension
227 activity lasted 1 h for each one.

228 Before sampling, quartz fibre filters (Ahlstrom Munksjo, Micro-quartz fibre paper MK306, Ø102
229 mm) were dried at 205 °C for 5 h and conditioned for 48 h at 20 °C and 50% relative humidity.
230 Blank filters were weighed three times every 24 h and kept in PETRI holders. After sampling,
231 filters were brought back to the laboratory and weighted after 24 and 48 h of conditioning at the
232 same temperature and humidity conditions. Filters were analysed for their pseudo-total
233 elemental content as soils to ensure the comparability of the measures. Microwave acid
234 digestion using 10 ml of *aqua regia* was performed using half filter in PTFE bombs (Ethos D,

235 Milestone). Resulting solutions were filtered on cellulose filters (Whatman Grade 5) and diluted
236 with ultrapure water to a final volume of 50 ml. Elemental contents were determined in all
237 samples using ICP-MS (NexION 350D, Perkin Elmer).

238 It was assumed that heavy metals content in PM₁₀ emitted under field conditions derives entirely
239 from soil and that the eventual contribution of the tractor combustion engine is negligible (Telloli
240 et al., 2014). Therefore, it was possible to relate the chemical analyses on the re-suspended
241 PM to the field measured PM₁₀ concentrations. The elemental concentration of TE in PM₁₀
242 (C_{dust} , $\mu\text{g g}^{-1}$) was then converted to elemental concentration in the air at 4 m distance from the
243 tractor (C_{air} , $\mu\text{g m}^{-3}$), by referring it to the overall PM₁₀ concentration measured in the field
244 (C_{PM10} , g m^{-3}) according to the following formula:

$$245 \quad C_{air} = C_{dust} C_{PM10}$$

246

247 2.6. Statistical analysis

248 A statistical analysis was conducted using the R software (R Core Team, 2019) to highlight
249 differences among EFs and ERs observed for rotary harrowing with different operational
250 parameters. For T1, the effect of tractor speed on EFs and ERs was tested (significance level
251 chosen was $\alpha < 0.05$) implementing a mixed model (*lme* procedure from *nlme* R package;
252 Pinheiro et al., 2018) to account for the nested experimental design. The model was set having
253 tractor speed (S) as a fixed factor and the plot as random factor. The distribution of within-
254 groups errors and random effects were graphically assessed to verify the model assumptions
255 (Pinheiro et al., 2006). The mean EF values were calculated (using *emmeans* R package;
256 Lenth, 2019) for each tractor speed level and post-hoc test comparison were performed
257 according to the Bonferroni post-hoc method (using the *multcomp* R package; Hothorn et al.,
258 2008).

259 For T2, the data were analysed through a mixed model (*lme* procedure from *nlme* R package;
260 Pinheiro et al., 2018) accounting for the nested effects included in the split plot layout of the
261 experiment. The model included, as fixed effects, the three operation parameters (bar
262 orientation, distance, and height) and their interaction. The random effects were distributed on
263 the three levels of the split plot, which included the following nested effects: height in distance,
264 distance in orientation and orientation in parcel.

265

266 **3. Results**

267 *3.1. Environmental conditions during the trials: soil characteristics and atmospheric* 268 *conditions*

269 In T1, wind speed and direction varied consistently during the tractor passages. The average
270 wind speeds were 0.81 ± 0.07 and 0.91 ± 0.07 m s⁻¹ at 2 and 4 m above ground, respectively.
271 Atmospheric stability condition was estimated to fall within the B PG_{class}, meaning that the
272 atmosphere was unstable during the essay. In T2, wind speed was slightly higher as compared
273 to T1 (1.46 ± 0.12 m s⁻¹ at 2 m and 1.7 ± 0.15 m s⁻¹ at 4 m). Atmospheric stability condition fell in
274 the PG_{class} C (slightly unstable atmosphere) for most passages, exception made for 6 of them,
275 for which the PG_{class} was B. The atmospheric stability condition of each passage was used for
276 modelling the EF. Windrose graphs illustrating the frequencies of main wind directions and
277 speeds during T1 and T2 are shown in Figure 4 and 5 (the graphs were obtained applying the
278 openair R package by Carslaw and Ropkins, 2012).

279 The low wind speed conditions observed are consistent with the annual average wind speed
280 reported for the Piedmont region by the regional ambient protection agency (ARPA Piemonte;
281 Fratianni et al., 2007).

282 Table 1 illustrates the main physico-chemical characteristics of the analysed soils. Both soils
283 were sub-acid and their total carbon content was similar and in line with their agricultural use,
284 as well as the other determined chemical characteristics. The moisture content of the soils
285 during the trial was 8.64 ± 0.03 % and 9.02 ± 0.02 %, on mass, in T1 and T2, respectively.
286 Although the finer texture of T2 soil as compared to T1 could lead to a higher emission potential,
287 it is speculative to draw conclusion on the base of texture information only, since it is known
288 that also soil aggregates stability can have a great impact on the final emissions (Madden et
289 al., 2010).

290 The overall environmental conditions observed during both T1 and T2, with coarse soil texture,
291 relatively high soil moisture content and low wind speed conditions, may lead, according to
292 previous studies, to relatively low emissions (Avecilla et al., 2017; Cassel et al., 2003; Madden
293 et al., 2010).

294

295 3.2. *EFs for rotary harrowing and effect of operational parameters on the emissions and*
296 *plume concentrations*

297 The results of the statistical analyses for T1 and T2 tilling events are summarized in Table 2. In
298 T1, the average EF for rotary harrowing with packer roller was $8.9 \pm 2.0 \text{ mg m}^{-2}$, considering
299 data gathered at all three speeds. The tractor speed was shown to alter significantly the EFs,
300 with the lower speed (S1) causing higher emissions compared to S2 and S3 (Table 2). On the
301 contrary, tractor speed had no significant effect on the ERs. In T2, the average EF for rotary
302 harrowing with packer roller was of $9.5 \pm 2.5 \text{ mg m}^{-2}$ (averaging all the trials). No significant effect
303 on the emissions was highlighted for bar height, bar orientation and bar distance nor their
304 interaction. From the obtained results, it appears that only bar distance could possibly affect
305 the emission, since the average EFs are generally higher in D1 as compared to D2 (Table 2),
306 although this effect is not statistically relevant. Some differences, although not significant
307 ($P > 0.05$), can be observed for different settings at distance D1, while practically no variation is
308 shown among EFs at distance D2.

309 Peak concentrations measured during tractor passes, at 4 m distance, were $641 \pm 40 \text{ } \mu\text{g m}^{-3}$ in
310 T1 and $3461 \pm 329 \text{ } \mu\text{g m}^{-3}$ in T2, averaging all the passages. The average downwind
311 concentration increases near the source (at 1 m height) are plotted in Figure 4 and 5, as
312 estimated for T1 and T2. Estimated PM_{10} concentration increments averaged between 12 (at
313 the field edge) and $0.1 \text{ } \mu\text{g m}^{-3}$ (at more than 300 m from the source). The plume in T2 appears
314 to be less horizontally spread as compared to T1 (due to the different stability conditions).
315 Downwind concentration in T2 also appears to be slightly higher. This was probably due both
316 to the reduced plume dispersion and to the higher wind speed registered that day.

317

318 3.3. *Elemental characterization of soils and soil-emitted PM_{10}*

319 The particle size distribution of the elements in soils has been demonstrated to be a key
320 parameter when exploring the possible risks associated to soil contamination (Ajmone Marsan
321 et al., 2008; Kong et al., 2012; Padoan et al., 2017). *Aqua regia* extractable concentrations of
322 elements in bulk soils are reported in Table 3. The TE contents in the resuspended PM_{10} fraction
323 is expressed in Table 3 as enrichment ratio, being the ratio between the content of each element
324 in the resuspended PM_{10} and in the bulk soil it originated from.

325 Both agricultural soils appeared as not contaminated (according to DLgs 152-2006), in
326 accordance with their long-term agricultural use. In soils, most of the elements appeared
327 enriched in the finer fraction. Only the major elements and, to some extent, the elements typical
328 of the parent material (such as Li, Sc, Co) had a similar concentration in both size fractions.

329 The enrichment in the finer fraction was particularly evident for Cu, Zn, Mo, Cd, Sn and Ba,
330 where the PM₁₀ soil had concentrations one order of magnitude higher than the bulk soil.

331 From the soil-related PM₁₀ fraction concentrations, we calculated the resulting concentration in
332 air according to the total concentration of atmospheric PM at 4m of distance from the source
333 (Table 4). From the results appear that the plume is enriched in TE, with point concentrations
334 of As, Cd and Ni higher than the legislation limits for PM₁₀ atmospheric pollution, respectively
335 6, 5 and 20 ng m⁻³ (MATTM, 2010). Although they refer to the annual average concentration,
336 punctual concentrations in the plume during T2 were up to 50 times the limit.

337

338 **4. Discussion**

339 The PM₁₀ emission factors calculated for rotary harrowing (8.9±2.0 in T1 and 9.5±2.5 mg m⁻² in
340 T2; averaged over all tractor passages) were found to be substantially lower as compared to
341 the one reported by Öttl and Funk (2007) for fixed-tooth harrow in Germany on a sandy soil
342 (83.3% of total sand), which was of 82 mg m⁻². The lower emissions observed are probably due
343 to the overall soil and environmental conditions and to the different implement used. The soil,
344 in both T1 and T2, had a high moisture content (8.64% in T1 and 9.02% in T2, on mass), being
345 in the range of the threshold levels of soil moisture, of 2 and 10 % on mass, over which,
346 according to Funk et al. (2008), very low PM emissions occur in sandy and silty soils
347 respectively. The lower sand content in the two Italian soils could have favoured an improved
348 soil structure and aggregation, which it is known to mitigate dust emissions (Madden et al.,
349 2009). The shielding structure which is present in rotary harrows and absent in fixed-tooth ones
350 could have important emission containment effect. Moreover, the wind speed registered during
351 the trial was lower than the 1.9 m s⁻¹ reported by Öttl and Funk (2007). Since the atmospheric
352 conditions registered during the trials are quite common in the Northwest of Italy (Fratianni et
353 al., 2007) and no previous assessments have been done for rotary harrows, the gathered EFs
354 could be considered as a first reference for this type of soil tillage.

355 A difference between EFs for low (S1), standard (S2) and high tractor speed (S3) was observed
356 during first trial. The emissions observed with S1 were in fact higher as compared to the other
357 treatments, which did not differ significantly among each other. This could be due to the longer
358 period in which the harrower insists on the same volume of soil when operating at the lower
359 speed. This explanation is further confirmed by the fact that no significant difference was
360 observed among the ERs generated at different speeds, meaning that the harrower emits the
361 same amount of PM₁₀ per second of work in each thesis. This indirect effect of tractor forward
362 speeds probably could apply only to the rotary harrower, which actively disturbs the soil, but
363 not for traditional soil tilling techniques, which have a passive action on the soil. Usaborisut and
364 Praserkan (2019), and Kushwaha and Linke (1996) have shown that an increase in the tractor
365 speed did not affect PM₁₀ releases during harrowing, although a reduction of soil fragmentation
366 should be obtained by raising the working speed. An increase in forward speed, in fact,
367 determines a stretching of the cycloid described by the harrow rotors tines with a consequent
368 increment of clods diameter (Raparelli et al., 2019).

369 In T2, altering bar height and orientation had no significant effects on PM₁₀ emissions. The
370 obtained results also showed a slight emission reduction while increasing the horizontal
371 distance between the levelling bar and the harrower. However, this effect was not statistically
372 relevant, and the bar distance could possibly affect the efficiency of the levelling bar itself. From
373 field observations it appeared that, when the bar was positioned at distance D2, the soil failed
374 to accumulate in correspondence of the harrower teeth. Therefore, to properly test this
375 mitigation opportunity, further trials should be carried out to better investigate its effect on
376 emissions but also on soil aggregates. It is important, when considering possible PM mitigation
377 options, to maintain the efficacy of the agricultural operation unaltered.

378 Peak concentration measured during trials near the tractor passes were $641 \pm 40 \mu\text{g m}^{-3}$ in T1
379 and $3461 \pm 329 \mu\text{g m}^{-3}$ in T2. The higher concentration during T2 as compared to T1 was
380 probably attributable to the higher wind speed, causing a more stable and focused plume, and
381 to wind direction, diagonal to the tractor movement, which permitted the operator to put the
382 sensor more in line with the plume centreline. The observed concentration levels were
383 consistent with those reported in previous studies (Moore et al., 2015; Clausnitzer and Singer,
384 1997) during land preparation activities. The main concerns related to those concentrations are
385 related to farmers' professional health risks. In fact, exposure to high levels of PM₁₀ in farming

386 environment can lead to severe health effects and possibly to fatal consequences (Moloczniak,
387 2002; Kirkhorn and Garry, 2000; Schenker, 2000). Although modern tractor cabins are provided
388 with technologies, such as air filters, to protect the operators from these risks, still a lot of
389 assessments are to be done to ensure a sufficient personal protection and to provide a safe
390 work environment for farmers.

391 Soil-related PM appeared to contain high concentrations of TE, especially those elements
392 deriving predominantly from anthropic sources. Elements such as Cu, Zn, Ni, Cr, Cd, Sn and
393 Ba had, in this fraction, concentrations higher than the legislation limits for soils, as observed
394 in previous studies on different soils (Padoan et al., 2017). This, in turn, affected atmospheric
395 concentrations of metals in the plume. These were calculated, for some of the regulated
396 elements in Italy (Ni, As and Cd), up to 50 times the limit for the annual average threshold levels
397 established by the legislation (MATTM, 2010). Although these were transient conditions, long
398 term exposition to such high concentrations could affect worker's health.

399 Modelled plume concentrations showed that PM₁₀ levels near the emission source can be
400 substantially affected from harrowing operations (Figure 4). In fact, PM₁₀ concentration
401 increases due to one hour of field harrowing, calculated at 100 m downwind of the source, were
402 estimated to be among 2 and 7 µg m⁻³ in both T1 and T2. Even if those concentration increases
403 may seem not too high at a first glance, it is important to take into account that land preparation
404 activities (as well as other cropping activities) are performed over extended cropped regions
405 (areas) and normally for several days or weeks. This concentration of agricultural emissions in
406 space and time is one of the main aspects that lead to sudden air pollution increases in rural
407 areas and nearby cities over specific year periods (Chen et al., 2017; Pavilonis et al., 2013).
408 Moreover, PM coming for agricultural operations can affect air concentrations and cause
409 relevant health effects even at medium and long-range distances (Hill et al., 2019).

410

411 **5. Conclusions**

412 The PM₁₀ EFs for rotary harrowing with levelling bar and for rotary harrowing combined with
413 packer roller were determined in low wind speed conditions. Since the atmospheric conditions
414 in which the trials have been made are quite common in Northwest of Italy (Fратиanni et al.,
415 2007) and no previous assessment have been done for rotary harrowing in North Italy, the

416 gathered EFs could be used as a first reference EFs for this type of soil tillage under moist soil
417 conditions. A further assessment should be performed to investigate the PM flux caused by the
418 same implement with drier soil conditions.

419 It was observed that lowering the tractor forward speed at 0.6 m s^{-1} has a negative effect on
420 PM emissions, causing them to increase significantly.

421 Major and trace elements in soil-generated PM_{10} were analysed, founding a higher content of
422 TE in the PM_{10} fraction than in the original soil sample, meaning that agricultural activities can
423 play a role in the transient increase of metals content in the atmospheric PM_{10} , even in regions
424 with low soil pollution.

425 Concentrations of PM_{10} at a distance of 4 m from the tractor passage were found to be up to
426 69 times higher than the daily limit fixed by WHO (2006), raising some concern for farmers
427 health. Estimated concentration raises near-source were also substantial (plus 2 to $7 \mu\text{g m}^{-3}$ at
428 100 m of distance from the emission source). Moreover, elemental (Ni, As and Cd)
429 concentration levels near the tractor path were also high, being several times higher than
430 regulatory threshold levels.

431 This first study highlighted the need of studies on the assessment of the emissions from
432 agricultural activities and to further investigate the effects of mechanic implements and
433 operative parameters on emission fluxes. Moreover, the dispersion of agricultural PM should
434 be assessed also including long-range transport, and focusing on the investigation of the
435 potential health impact of the contaminants present in soil particulates.

436

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446

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619 **List of tables**

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625 Table 3. Concentrations (\pm standard error) of each element determined in the bulk soils and
626 enrichment ratio in soil-derived PM₁₀.

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628 Table 4. Air elemental concentrations at 4 m distance from the tractor.

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640 Table 1.

Soil characteristics	T1	T2
Coarse sand (%)	23	9
Fine sand (%)	33	31
Coarse silt (%)	12	14
Fine silt (%)	26	28
Clay (%)	6	18
pH (H ₂ O)	6.1	6.2
TC (%)	1.34	1.10
TN (%)	0.18	0.18
Soil density (g cm ⁻³)	1.5	1.4

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Table 2.

Trial	Operative parameters			Mean EF (mg m ⁻²)	SE	P _{value}
T1	Forward speed					
		S1		13.4 ^a	2.1	0.002
		S2		3.6 ^b	1.6	
		S3		4.8 ^b	2.0	
T2	D1	Distance	Height	Orientation		0.060
			H	S	33.7	
		L	S	12.4	9.0	
		H	I	5.0	7.3	
		L	I	14.9	7.3	
	D2	H	S	5.0	3.3	
		L	S	6.1	3.3	
		H	I	5.5	4.4	
L		I	3.8	4.4		

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673 Table 3.

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		T1		T2	
		Bulk soil 1	Enrichment ratio in PM ₁₀ fraction	Bulk soil 2	Enrichment ratio in PM ₁₀ fraction
Mg	%	0.95 ± 0.02	1.5	1.08 ± 0.04	1.7
Al		3.73 ± 0.02	1.0	3.05 ± 0.21	1.5
K		0.99 ± 0.04	1.0	0.67 ± 0.09	1.5
Ca		0.80 ± 0.07	3.1	0.47 ± 0.03	2.7
Fe		1.24 ± 0.01	2.0	4.36 ± 0.11	0.5
Li	(µg g ⁻¹)	50 ± 4	0.8	42 ± 2	1.3
Sc		13 ± 1	1.5	8.8 ± 1.3	1.6
V		86 ± 18	1.8	66 ± 5	1.6
Cr		189 ± 25	6.3	91 ± 24	5.0
Mn		1758 ± 110	1.5	1225 ± 19	1.5
Co		22 ± 1	2.2	20 ± 2	1.6
Ni		135 ± 11	4.4	84 ± 20	3.1
Cu		62 ± 5	7.3	66 ± 5	4.1
Zn		104 ± 8	7.8	58 ± 5	9.3
As		14 ± 1	0.9	13 ± 1	1.7
Sr		75 ± 3	2.8	34 ± 10	2.4
Mo		4.9 ± 3.7	21	< 0.05	
Cd		0.36 ± 0.05	39	0.13 ± 0.03	94.3
Sn		2.8 ± 0.2	13	2.6 ± 0.2	4.6
Sb		0.58 ± 0.25	20	0.58 ± 0.02	5.2
Ba		246 ± 6	5.5	86 ± 10	7.3
Pb		37 ± 0.4	2.5	27 ± 1	1.7

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680 Table 4.

		T1	T2
Mg	$\mu\text{g m}^{-3}$	12	62
Al		29	157
K		6.4	34
Ca		8.1	44
Fe		14	76
Mn		1.2	6.2
Li	ng m^{-3}	34	183
Sc		9	50
V		67	363
Cr		294	1585
Co		21	111
Ni		168	906
Cu		176	949
Zn		345	1865
As		14	75
Sr		52	283
Mo		64	347
Cd		8	43
Sn		8	41
Sb		2	10
Ba		401	2168
Pb		29	154

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689 **List of figures**

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692

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696

697 Figure 4. a) Windrose illustrating the frequencies of main wind directions and speeds during
698 T1; b) Estimated downwind concentration increases near source (area of 1 ha) during T1..

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700 Figure 5. a) Windrose illustrating the frequencies of main wind directions and speeds during
701 T2; b) Estimated downwind concentration increases near source (area of 1 ha) during T2.

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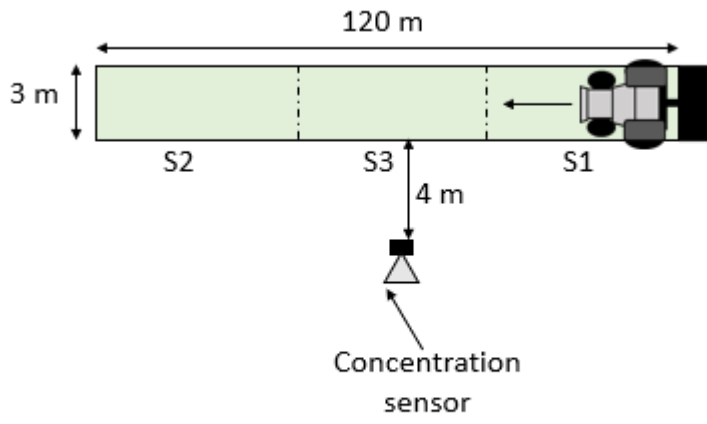
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713 Figure 1.



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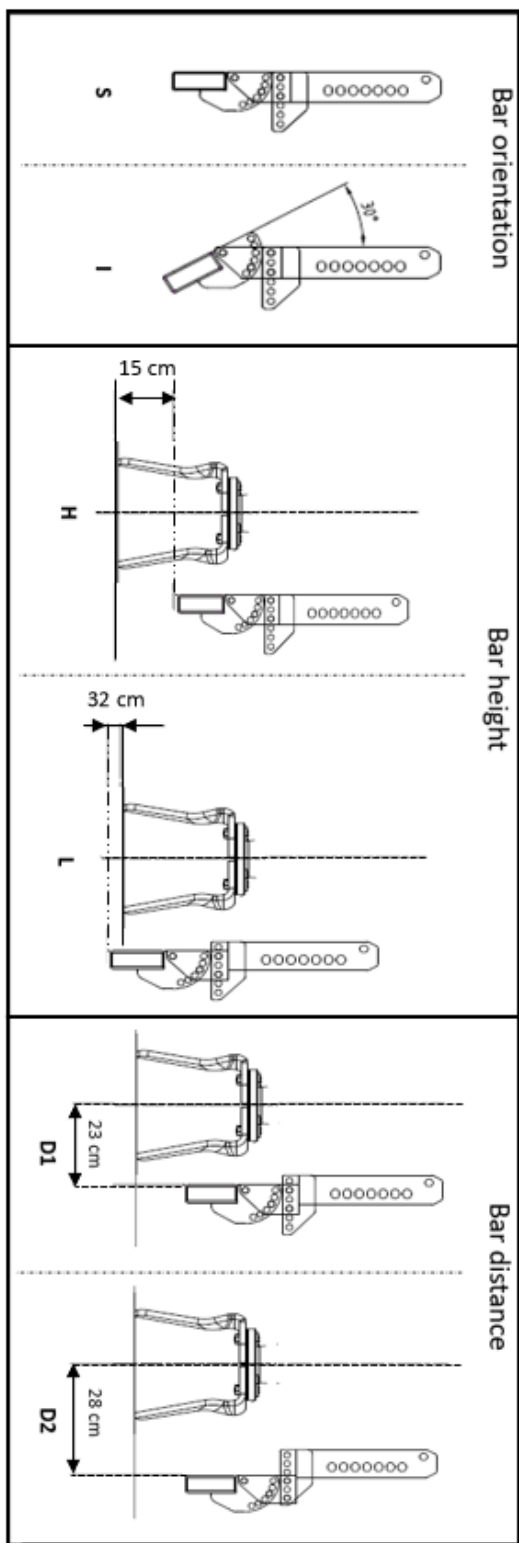
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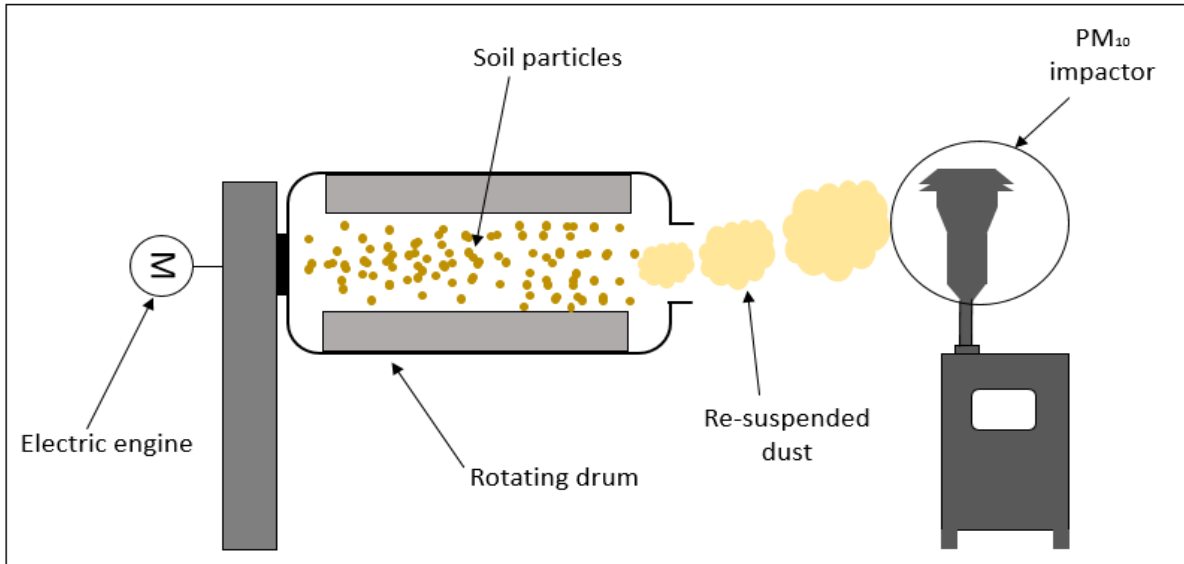
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Split plot design

Parcel	Factor	Level	Parcel	Factor	Level
Parcel A	H	L	L	H	D1
		L			D2
	L	L	H	H	D1
		L			D2
	H	L	L	H	D1
		L			D2
L	L	H	H	D1	
	L			D2	
Parcel B	H	L	L	H	D1
		L			D2
	L	L	H	H	D1
		L			D2
	H	L	L	H	D1
		L			D2
L	L	H	H	D1	
	L			D2	
Parcel C	H	L	L	H	D1
		L			D2
	L	L	H	H	D1
		L			D2
	H	L	L	H	D1
		L			D2
L	L	H	H	D1	
	L			D2	

732 Figure 3.



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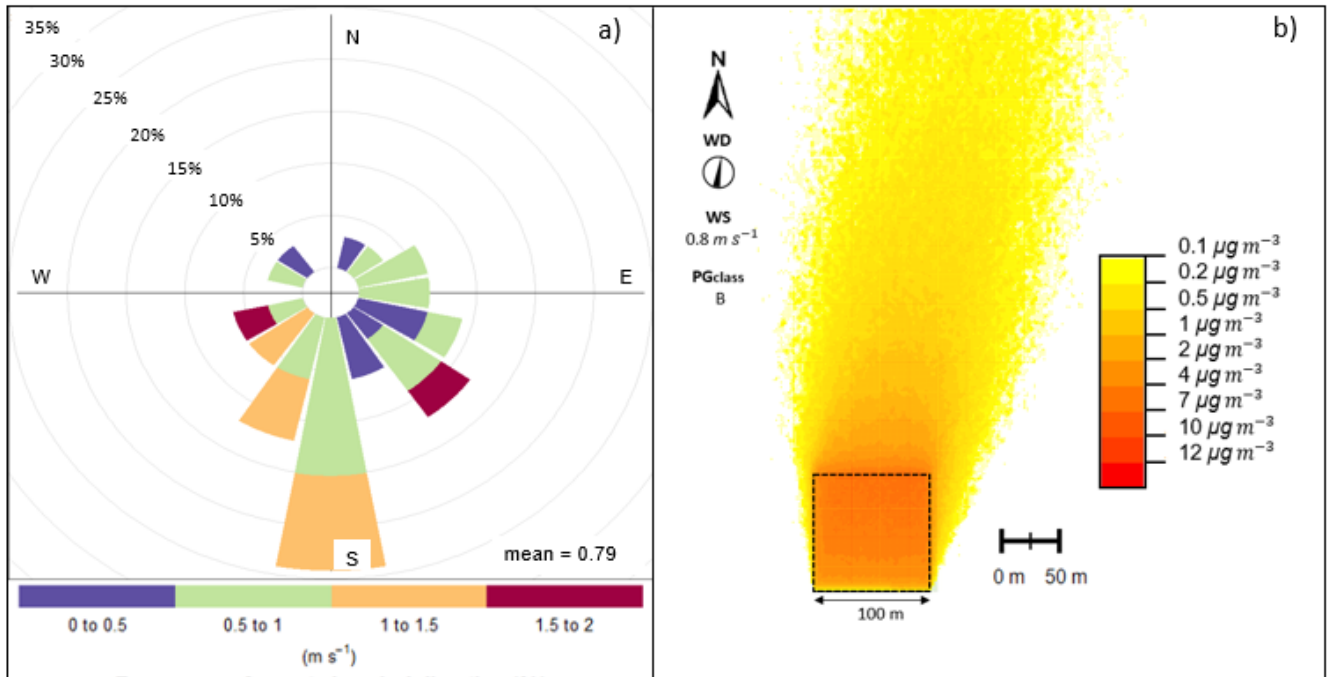
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748 Figure 4.

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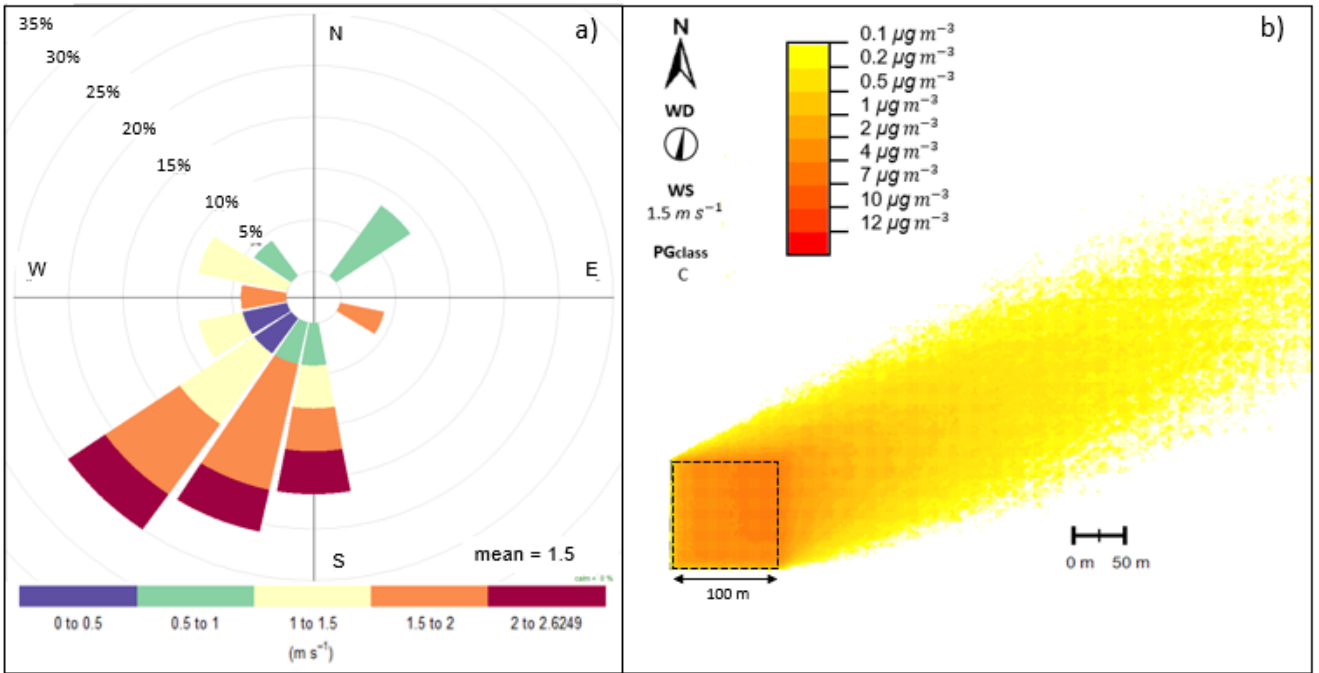
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Figure 5.



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