

1 The increase of rainfall erosivity and initial soil erosion processes due to rainfall 2 acidification

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13 14 15 Abstract

16 The drastic growth of population in highly industrialized urban areas, as well as fossil fuel use, are
17 increasing levels of airborne pollutants and enhancing acid rain. In rapidly developing countries
18 such as Iran, the occurrence of acid rain has also increased. Acid rain is a driving factor of soil
19 erosion due to the destructive effects on biota and aggregate stability; however, little is known
20 about its impact on specific rates of erosion at the pedon scale. Thus, the present study aimed to
21 investigate the effect of acid rain at pH levels of 5.25, 4.25 and 3.75 for rainfall intensities of 40,
22 60 and 80 mm h⁻¹ on initial soil erosion processes under dry and saturated soil conditions using
23 rainfall simulations. The results were compared using a two-way ANOVA and Duncan tests and
24 showed that initial soil erosion rates with acidic rain and non-acidic rain under dry soil conditions
25 were significantly different. The highest levels of soil particle loss due to splash effects in all
26 rainfall intensities were observed with the most acidic rain (pH= 3.75), reaching maximum values
27 of 16 g m⁻² min⁻¹. The lowest levels of particle losses were observed in the control plot where non-
28 acidic rain was used, with values ranging from 3.8 to 8.1 g m⁻² min⁻¹. Similarly, under saturated
29 soil conditions, the lowest level of soil particle loss was observed in the control plot and the highest
30 peaks of soil loss was observed for the most acidic rains (pH= 3.75 and pH= 4.25), reaching

31 maximum average values of $40 \text{ g m}^{-2} \text{ min}^{-1}$. However, for saturated soils with acidic water, but
32 with non-acidic rain, the highest soil particle loss was observed for the control plot for all the
33 rainfall intensities. In conclusion, acidic rain has a negative impact on soils, which can be more
34 intense with a concomitant increase in rainfall intensity. Rapid solutions, therefore, need to be
35 found to reduce emission of pollutants into the air, otherwise, rainfall erosivity may drastically
36 increase.

37 **Keywords:** acid rain; splash erosion; runoff; soil loss

38

39 **1. Introduction**

40 The increasing use of fossil fuels in industry, manufacture and transport, especially in rapidly
41 developing countries such as Iran, is emitting a large quantity of pollutants into the atmosphere
42 and causing damage to human health and to ecosystems (Ashtari et al., 2018; Bahrami Asl et al.,
43 2018; Mohammadiha et al., 2018). Moreover, due to the rapid growth of urbanization and the
44 concentration of population close to industrial areas such as Tehran, Isfahan, Arak, Ahvaz and
45 Mashhad, the development of old and worn transportation vehicles, factories, and power plants
46 that use fossil fuels, air pollution has increased (Fanni, 2006; Hafeznia et al., 2017; Yousefi et al.,
47 2017). However, in polluted cities in developing countries, Atash (2007) highlighted that the
48 implementation of 10-year strategic plans has been delayed. In recent years, air pollution rates
49 have exceeded dangerous levels which forced the authorities to enforce traffic constraints and
50 school closures. In a study performed by Lelieveld et al. (2015), Tehran was ranked among the
51 cities with a high mortality rate due to human exposure to long-term air pollution. According to
52 Shahbazi et al. (2016), annual pollution from fixed sources such as factories, and mobile industries

53 including the transportation agency in Tehran for 2013, amounted to 37.4 kg, 85.5, 506.7, 83.6 and
54 8.5 kg for SO_x, NO_x, CO, VOCs and PM emissions, respectively. Also, Alizadeh Choobari et al.
55 (2016) investigated PM₁₀ and PM_{2.5} levels in the north of Tehran, which were found to be
56 significantly higher than the national average on a per m² basis. The excessive emission of
57 pollutants and acidifying compounds such as nitrous oxides and sulphuric acid into the atmosphere
58 favours conditions for acid rain in the form of fog, snow and rain (Uchiyama et al., 2017; Zheng
59 and Yu Hong, 1994). Acid rain is defined as a rain characterized by pH values lower than 5.6
60 (Mirhosseini et al., 2009; Purohit, and Kakrani, 2002; Neill, 1993; Welburn, 1990). Acid rain
61 adversely affects ecological and environmental processes (Wang et al., 2018; Wei et al., 2017).
62 Over the last 50 years, several researchers have highlighted the negative environmental impacts of
63 acid rain, including destruction of buildings and tools (Yokom and Beer, 1983), forest and
64 geological formation degradation (Ulrich, 1980, Driscoll et al., 2001), and adverse effects on crops
65 and soil fertility (Ferenbaugh, 1976; Pell et al., 1987; Irving, 1987).

66 Evidence is gathering that this acid rain is increasing close to highly populated and industrial areas
67 in Iran. For example, Moarref et al. (2011) recorded pH values of about 2.4 by evaluating the
68 chemical composition of rain in Ahwaz city. Mirhosseini et al. (2009) investigated the occurrence
69 of acid rain in Sarcheshmeh area, located in the Kerman province, and determined that that
70 occurrence is an inevitable phenomenon close to highly industrial and populated regions.
71 However, these authors remarked that there is a lack of quantification of the effects of acid rain on
72 soil processes such as water soil erosion (Mirhosseini et al., 2009; Moarref et al., 2011). As acid
73 rain impairs soil fertility and vegetation development, the areas with bare soil surfaces will increase
74 (Driscoll et al., 2001). Lack of vegetation is one of the most important driving factors of soil
75 erosion processes in watersheds and hillslopes (Cerdà et al., 2018; Saleh et al., 2017; Parsakhoo et

76 al., 2012a). Water soil erosion is initiated by the collision of rain drops with the soil surface
77 (Ellison, 1944; Ellison, 1947; Free 1952). Rain drops can separate and move soil particles and
78 aggregates (Fernández-Raga et al., 2017; Marzen et al., 2015).

79 The splash effect on soils is the first stage in the erosion process, which is the result of
80 bombardment of the soil surface by rain droplets (Qinjuan et al., 2008, Wuddivira et al., 2009).

81 The splash effect, by crushing soil particles and reducing their diameter, leads to a reduction of
82 soil particle resistance against transport, and also a decrease in water penetration in soil surface
83 layers, which in turn can increase runoff, erosion and sediment transport (Barry et al., 2010;
84 Sadeghi et al., 2017).

85 However, there is a lack of information about the degree of disintegration of soil aggregates due
86 to splash erosion at different levels of rain acidity (Xu et al., 2002; Manahan, 2005). A number of
87 negative effects of acid rain and soil erosion can occur, such as leaching of nutrient cations, release
88 the toxic elements and soil acidification (Mirhosseini et al., 2009). Also, the sedimentation of the
89 elements in the form of insoluble hydroxides and carbonates and organic complexes can increase.
90 Moreover, when soil acidification occurs, heavy metals such as zinc and cadmium are mobilized,
91 potentially leading to toxic concentrations (Smith, 1994).

92 Analysis of initial soil erosion processes in combination with acid rain is time-consuming and the
93 intra-plot variation is high. Therefore, one of the most direct measurement methods of initial soil
94 erosion, where rainfall conditions can be controlled, is the rainfall simulator (Iserloh et al., 2013),
95 which has been widely used in Iran (Parsakhoo et al. 2012b; Kavian et al. 2014; Safari et al. 2016;
96 Sadeghi et al., 2017; Ayoubi et al., 2018).

97 The main aim of this study was to assess the impact of acidic rain at different pH levels and
98 intensities on initial soil erosion processes under distinct soil conditions using a small portable

99 rainfall simulator. The experiments were conducted under laboratory conditions to avoid any
100 external factors such as wind, soil property changes or inclinations. We hypothesize that this first
101 approach will allow us to show if acidic rain *per se* directly affects initial soil erosion processes,
102 as a basis for future research under natural conditions and in different environments.

103 **2. Materials and methods**

104 **2.1. Soil sampling**

105 The altitudinal range of the case study region is between 150 and 300 m above sea level with a
106 mean annual precipitation of 600 mm. Most events occurs are during the winter and spring seasons
107 (November–May), with a means annual temperature of 18.5 °C. Two hundred and fifty kg of soil
108 was collected from a typical cultivated field where the main tillage practice is wheat dry farming
109 in Miandorood region, in the range of 36° 33' to 36°35' latitude and 53° 10' to 53°13' longitude
110 close to Sari (the capital of Mazandaran province) from a surface soil to a depth of 0-20 cm,
111 following the recommendations of Angulo-Martinez et al. (2012) for laboratory experiments.
112 Samples were transported to the laboratory and sieved using a 2 mm sieve. They were then dried
113 in an oven at 105°C for 24 hours before rainfall simulations (Mohammadi and Kavian et al. 2015),
114 in order conserve the same moisture conditions in all samples.

115 Pedogenesis was generated on marl and unconsolidated sand deposits of the Pliocene. In general
116 slopes are gentle; lower than 10%. The soil is characterized by a loam texture (with silt, sand and
117 clay components as 48.6%, 33.8% and 17.6%, respectively), a low organic matter content of 1.2%,
118 electrical conductivity of 0.499 dsm⁻¹, calcium carbonate content of 29.3% and a pH value of 7.36.
119 After simulating the acid rain, organic matter and calcium carbonate of the soil were evaluated in
120 some samples.

121

122

123 **2.2. Rainfall simulator characteristics**

124 The rainfall simulator was mounted on a metal A- frame structure. The height can range from 2 to
125 2.7 m depending on the purpose. The telescopic legs allow the height of the nozzles to be changed,
126 which regulates the rainfall intensity and kinetic energy. The rainfall simulator can be used on
127 rugged terrain as the telescopic legs allow levelling at any slope angle from 0 to 45°. Simulated
128 rainfall is produced with two movable Veejet 80100 nozzles (Pall et al., 1983; Blanquies et al.,
129 2003; Chouksey et al., 2017) with a diameter of 4.5 mm. Each nozzle is installed in a metal deposit
130 to collect and reuse the excess of rainfall (that is not sprayed on the plot), and then returned to the
131 pumping system. The rainfall simulator was used in the laboratory of the Sari Agricultural Sciences
132 and Natural Resources University. The distilled water is pumped to the nozzles by means of a
133 flexible hose with 15-mm diameter connected to an electric pump. The water pressure is monitored
134 by a barometer installed in the transfer hose that allows the pressure to be regulated between 0 and
135 160 KPa (Fig. 1). A control board was designed with a programming capability of ten precipitation
136 programs, to perform experiments with different rainfall characteristics. The control board can be
137 used to set the velocity fluctuation nozzles, the oscillation angle of nozzles from 0° to 60°, and the
138 duration of each rainfall event from 1 min to 1 hour. The plot size is 0.5 * 1 m² and 3 splash cups
139 are located in the plot (figure 2). Splash cups were adapted from Morgan's original design (Morgan,
140 1978).

141 Soil splash rates in eleven different treatments of rainfall acidity with dry and saturated soils, at
142 three intensities of 40, 60 and 80 mmh⁻¹, based on the average rainfall in three replicates, were
143 recorded (Table 1). A total of 99 samples were tested.

144 Figure 2 shows the schematic representation of splash cups considering Morgan's original design
145 to calibrate kinetic energy and rainfall intensity (Morgan, 1978). The rainfall simulator is able to
146 generate drops with a diameter from 0.2 to 9.9 mm. The fall velocity varied from 0.8 to 9.2 m s⁻¹
147 for different diameter classes in the height of 0.5 m above the soil surface. The minimum and
148 maximum droplet sizes decreased by increasing the operating pressures from 20 to 80 kPa. The
149 drop diameter of the simulated rainfall is close to natural precipitation in the region of Miandorood
150 (Kavian et al., 2018).

151 Simulated acid rainfall contained sulfuric acid and nitric acid with a ratio of 2:1 using the volume-
152 concentration formula, which was simulated at three pH levels: 3.75, 4.25 and 5.25. These levels
153 were selected because they are to the most representative ranges found in several recent studies
154 with negative effects in agricultural or natural fields and buildings (Livingston, 2016; Du et al.,
155 2017; Mahdikhani et al., 2018; Zeng et al., 2018). Moreover, rainfall simulations with non-acidic
156 rain (pH= 7.53) was also conducted in order to compare acid rain with a control situation.

157

158 **2.3. Experimental procedure**

159 Soil samples were located in the splash cups under dry and saturated conditions, using firstly non-
160 acidic water and then acidic water with pH values of 3.75, 4.25 and 5.25. After 10 minutes of
161 simulated rainfall, the splashed soil particles inside the splash cups after each treatment and
162 intensity were separated in different containers at the end of the experiments. The yielded particles

163 were air-dried for 24 hours (Kavian et al., 2014). After the extra water was drained, the remaining
164 sediment was transferred into suitable containers of specified weight and were dried in an oven at
165 105 °C for 24 hours, then weighted using a scale (Sutherland & Zieglers, 1998).

166 Finally, the splash erosion rate was calculated using equation 1 (Qinjuan et al., 2008).

$$167 \quad S = \frac{D_{t_2} - D_{t_1}}{(t_2 - t_1)A} \quad (\text{Eq. 1})$$

168 S = splash rate during the specified rainfall ($\text{gm}^{-2}\text{minute}$)

169 D_{t_1} = Soil weight before splash experiment (g)

170 D_{t_2} = Soil weight after splash test (g)

171 $\Delta t = (t_2 - t_1)$ rainfall duration (min)

172 A = area of splash cup (m^2)

173

174 **2.4. Statistical Analysis**

175 The normality of data was assessed using by Kolmogorov-Smirnov test at a significance level of
176 0.05. Comparison of means was carried out by one-way ANOVA and the interactive effects of the
177 factors were analysed using two-way ANOVA. Duncan's multiple range test was applied for
178 multiple mean comparisons at a significance level of 0.05. All statistical analyses were conducted
179 using SPSS 23.0 (IBM, USA).

180

181 **3. RESULTS AND DISCUSSION**

182 3.1 Comparison of drop splash effects on soil loss under dry conditions

183 Table 2 shows the results of one-way ANOVA of the different treatments at intensities of 40, 60
184 and 80 mmh⁻¹ for the dry soil. Significant differences for the different treatments for an intensity
185 of 40 mmh⁻¹ at a confidence interval of 95% and an intensity of 60 mmh⁻¹ at a confidence level of
186 99% are obtained. However, no significant differences are found for splash results with an intensity
187 of 80 mmh⁻¹. Considering the significant difference between the treatments at 40 mmh⁻¹, Duncan's
188 multiple range test was performed to show the difference among treatments. Figure 3 shows total
189 initial soil erosion results and Duncan's multiple range test for control plot and acidic rain
190 simulation under dry soil conditions. The highest particle loss due to splash effect in all degrees of
191 rainfall intensities are observed with the most acidic rain (pH= 3.75), reaching maximum values
192 of 16 g m⁻² min⁻¹. On the contrary, the lowest particle losses are obtained in the control plot, where
193 non-acidic rain is used with values ranging from 3.8 to 8.1 g m⁻² min⁻¹. The results show for 40
194 mm h⁻¹ that the AR1DS treatment is different (a) from the other three treatments, classified in
195 group b. For a rainfall intensity of 60 mmh⁻¹, the treatments are classified into three groups. The
196 most acidic rain (AR1DS treatment) is in the first group (a) again. AR2DS and AR3DS appear in
197 the same group (b) giving similar particle loss results, and the control plot (NARDS) is in group
198 c. Finally, for 80 mm h⁻¹ of rainfall intensity, the highest and the lowest values for splash were
199 observed in the most acidic rain (AR1DS). The results are very similar with no significant
200 difference among treatments at this high rainfall intensity, which even without acid rainfall, is able
201 to contribute in bare soils to a high soil particle loss values (Beguería et al., 2015; Eldridge and
202 Greene, 1994). Therefore, under dry conditions, we demonstrated that a higher concentration of
203 acid rain is able to increase soil erosion rates. The low rates observed for the control plot and the
204 lowest acidic rain (pH<5.25) could be due to the presence of calcium carbonate in our soil samples

205 (25.3%), which was functioning as a stabilizing factor for aggregate stability (Bakhshipour et al.,
206 2016). Therefore, parent material such as limestones or marls can act as a driving factor if
207 carbonates are correctly transferred to soil horizons, as Cerdà (2002) showed in Mediterranean
208 areas. However, when acid rain occurs, the lime present into the soils is neutralized; subsequently,
209 soil particles can be more easily separated (Gratchev and Towhata, 2016).

210

211 **3.2 Comparison of drop splash effects on soil loss under saturated conditions**

212 Table 2 also shows the results of the ANOVA test for soils under saturated conditions for the
213 intensities of 40, 60 and 80 mmh⁻¹. For 40 mmh⁻¹, there were no significant differences, despite
214 the increasing trend in soil particle loss values of acid rainfall compared to non-acidic rain, though
215 an increasing trend with more acidic rainfall was recorded. A significant difference in soil particle
216 loss is found for treatments with rainfall intensities of 60 and 80 mmh⁻¹.

217 Average values of total soil particle losses are shown in figure 4. Under saturated soil conditions,
218 the smallest amount of soil particle loss was registered in the control plot, as also seen under dry
219 soil conditions. The highest peaks of soil loss were obtained for the most acidic rains (pH= 3.75
220 and pH= 4.25), reaching maximum average values of 40 g m⁻² min⁻¹. However, for the Duncan
221 test, the differences are not significant at 40 mm h⁻¹, and only for 60 mm h⁻¹ the most acidic rainfall
222 shows a significant difference (AR3SS3). However, for 80 mm h⁻¹, a significant difference was
223 clearly seen among treatments, with the AR1SS1 treatment showing only slightly lower levels of
224 soil particle loss than the AR2SS2 treatment. These findings confirm the importance of previous
225 soil moisture in the plot. As other authors have found, saturated soils respond according to a
226 Hortonian model, where runoff is able to activate soil loss when soil is saturated (Gabarrón-

227 Galeote et al., 2013; Imeson and Lavee, 1998). Therefore, it is very important to pay attention to
228 soil water content prior to performing the rainfall experiments, because this can impact loss rates
229 (Hébrard et al., 2006; Wei et al., 2007). Kanga (1999) argued that when rainfall droplets hit the
230 ground the soil particles are disintegrated, and then, due to the water content and their reduced
231 adhesion force, particles are returned from the surface by droplets and soil particle loss increases.
232 Our results confirm the contention that in arid and semiarid areas, soil water content is able to
233 determine initial soil erosion rates, and an increase in acidification of the rainfall may also enhance
234 these rates. This is an important finding which will affect many regions, particularly for calcareous
235 soils in Iran. The rapid growth of urbanization and the increase in population close to large and
236 industrial Iranian cities are increasing emissions of pollutants to the air (Fanni, 2006; Hafeznia et
237 al., 2017; Yousefi et al., 2017), therefore, rapid solutions need to be found in order to stop this
238 confirmed problem. As other researchers in Iran have found, there are other driving factors that
239 also enhance soil erosion such as wind, bare soils and extreme rainfall events (Samani et al., 2016).
240 This new factor, acidic rain, needs to be added to the list of potential drivers of soil erosion (Tarolli,
241 2016). Therefore, solutions or regulations to limit air pollution should be implemented in order to
242 minimize, or stop, this confirmed land degradation process (Hansen et al., 2013; Smith et al.,
243 2008).

244

245 **3. 3. Evaluation of the effects of non-acidic rainfall on acidified saturated soil at different** 246 **rainfall intensities**

247 The negative effects of acidic rain in calcareous soils are clear; however, in order to observe an
248 inverse effect, the influence of non-acidic rain on acidic soils, we also conducted rainfall
249 simulations on saturated soils at three different pH values, 3.75, 4.25, 5.25, with three different

250 rainfall intensities (40, 60 and 80 mm h⁻¹) but with non-acidic rain. Table 2 shows the results of
251 one-way ANOVA for each treatment. There is no significant difference between the mean splash
252 rate of NARSS, NARSS1, NARSS2, NARSS3 at intensities of 40 and 60 mmh⁻¹, but there is for
253 80 mmh⁻¹ at the 99% confidence level. Figure 5 shows the comparison among each treatment at
254 three rainfall intensities (40, 60 and 80 mmh⁻¹) and pH levels using the Duncan's test at confidence
255 interval of 95%. For a rainfall intensity of 40 mm h⁻¹ and 60 mm h⁻¹, there were no significant
256 differences among treatments, registering the highest soil particle loss (17.1 g m⁻² min⁻¹) in the
257 control plot and the lowest one (10 g m⁻² min⁻¹) for pH values of 5.25. With a rainfall intensity of
258 80 mmh⁻¹, the control plot (NARSS) shows significant differences from the other treatments. The
259 lowest soil particle loss was found for the pH value of 5.25 (18.7 g m⁻² min⁻¹) with maximum rates
260 in the control plot (32.9 g m⁻² min⁻¹). This study shows the impact of soil saturation on increasing
261 soil particle loss.

262 According to the comparison of means, the results showed that the lowest splash rate was attributed
263 to the NARDS treatment. When comparing the average splash rate at the intensities of 40, 60 and
264 80 mmh⁻¹, it was observed that splash rate increases with increasing intensity, confirming the
265 findings of other studies (Gholami et al., 2016; Sadeghi et al., 2015); this means that with
266 increasing rainfall intensity, the erosion rate increases, which can be due to the fact that with
267 increasing rainfall intensity, the number of droplets that can hit the soil increases, and raindrops
268 can increase in size and as a result their mass and falling velocity increase, which leads to more
269 kinetic energy (Mohammadi and Kavian., 2015). The energy of the raindrops is a major factor in
270 the disintegration of soil aggregates (Valettea et al., 2006; Barry et al., 2010; Brodowski et al.,
271 2013), which can have a great influence on the separation of soil aggregates and results in splash
272 erosion (Liu et al., 2016).

273 Also, changes in soil organic matter under short term acidic rainfall are plausible. As we observed
274 in table 3, on dry and saturated soils, organic matter decreased after acidic rain from 1.24% to
275 0.97% and from 1.28% to 0.99%, respectively. Thus, further research is needed to investigate the
276 loss of soil we are registering because of the dissolution of organic matter, but now over long-term
277 periods.

278

279 **3.4. Challenges and further investigations**

280 Over long-time periods, acid rain might degrade vegetation and modify microbial communities
281 (Ling et al., 2010; Wu et al., 2016), both of which could conceivably change erosivity over time
282 (Xiao et al., 2017). However, we acknowledge that in this research using rainfall simulations, this
283 plausible mechanism for long term changes in erosivity cannot explain the findings presented here.
284 Splash erosivity is largely a physical process (Jomaa et al., 2012) and all of the studied soils are
285 similar, so the only difference can be in the way acidity affects the characteristics of the rain, for
286 example, by greater density or lower surface tension generating larger drops. It is widely accepted
287 that the mechanism cannot operate through impacts of acidity on the soil, as splash erosion is
288 caused when the water hits the soil, before any effect of the greater acidity can take place.
289 Therefore, future research lines must be conducted in order to develop a plausible and empirically-
290 verifiable mechanism to assess: why acid in the rain would increase erosivity instantaneously when
291 applied to the same soils, and, the specific pedological mechanism; or, ii) density, surface tension,
292 droplet size (and any other physical characteristic that might be relevant) for the acid amended
293 waters.

294

295 4. CONCLUSIONS

296 In this research, we tested the possible negative impact of acidic rain on initial soil erosion rates
297 in carbonate soils. Our research confirmed that the highest soil particle loss due to splash effect
298 across all rainfall intensities tested was observed with the most acidic rain (pH= 3.75) with the
299 lowest particle loss rates in the control plot, where non-acidic rain was used. A similar pattern was
300 seen for saturated soils. However, for acidic saturated soils, but with non-acidic rain, the highest
301 soil particle loss was seen in the control plot for all rainfall intensities. We demonstrated the
302 negative impact of acidic rain on soils, which could be exacerbated by a concomitant increase in
303 rainfall intensity. The development of rapid solutions and regulation by governments and land
304 planners is necessary to reduce emissions of pollutants to the air, because rainfall erosivity may
305 drastically increase.

306 We propose that further research must be conducted in order to develop a plausible and
307 empirically-verifiable mechanism to assess why acid in the rain would increase erosivity
308 instantaneously when applied to the same soils, or which rainfall characteristic is modified and is
309 able to destroy soil aggregates.

310

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