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

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Abstract

The drastic growth of population in highly industrialized urban areas, as well as fossil fuel use, are increasing levels of airborne pollutants and enhancing acid rain. In rapidly developing countries such as Iran, the occurrence of acid rain has also increased. Acid rain is a driving factor of soil erosion due to the destructive effects on biota and aggregate stability; however, little is known about its impact on specific rates of erosion at the pedon scale. Thus, the present study aimed to investigate the effect of acid rain at pH levels of 5.25, 4.25 and 3.75 for rainfall intensities of 40, 60 and 80 mm h⁻¹ on initial soil erosion processes under dry and saturated soil conditions using rainfall simulations. The results were compared using a two-way ANOVA and Duncan tests and showed that initial soil erosion rates with acidic rain and non-acidic rain under dry soil conditions were significantly different. The highest levels of soil particle loss due to splash effects in all rainfall intensities were observed with the most acidic rain (pH= 3.75), reaching maximum values of 16 g m⁻² min⁻¹. The lowest levels of particle losses were observed in the control plot where non-acidic rain was used, with values ranging from 3.8 to 8.1 g m⁻² min⁻¹. Similarly, under saturated soil conditions, the lowest level of soil particle loss was observed in the control plot and the highest peaks of soil loss was observed for the most acidic rains (pH= 3.75 and pH= 4.25), reaching
maximum average values of 40 g m\(^{-2}\) min\(^{-1}\). However, for saturated soils with acidic water, but
with non-acidic rain, the highest soil particle loss was observed for the control plot for all the
rainfall intensities. In conclusion, acidic rain has a negative impact on soils, which can be more
intense with a concomitant increase in rainfall intensity. Rapid solutions, therefore, need to be
found to reduce emission of pollutants into the air, otherwise, rainfall erosivity may drastically
increase.

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

1. Introduction

The increasing use of fossil fuels in industry, manufacture and transport, especially in rapidly
developing countries such as Iran, is emitting a large quantity of pollutants into the atmosphere
and causing damage to human health and to ecosystems (Ashtari et al., 2018; Bahrami Asl et al.,
2018; Mohammadiha et al., 2018). Moreover, due to the rapid growth of urbanization and the
concentration of population close to industrial areas such as Tehran, Isfahan, Arak, Ahvaz and
Mashhad, the development of old and worn transportation vehicles, factories, and power plants
that use fossil fuels, air pollution has increased (Fanni, 2006; Hafeznia et al., 2017; Yousefi et al.,
2017). However, in polluted cities in developing countries, Atash (2007) highlighted that the
implementation of 10-year strategic plans has been delayed. In recent years, air pollution rates
have exceeded dangerous levels which forced the authorities to enforce traffic constraints and
school closures. In a study performed by Lelieveld et al. (2015), Tehran was ranked among the
cities with a high mortality rate due to human exposure to long-term air pollution. According to
Shahbazi et al. (2016), annual pollution from fixed sources such as factories, and mobile industries
including the transportation agency in Tehran for 2013, amounted to 37.4 kg, 85.5, 506.7, 83.6 and 8.5 kg for SO\textsubscript{X}, NO\textsubscript{X}, CO, VOCs and PM emissions, respectively. Also, Alizadeh Choobari et al. (2016) investigated PM\textsubscript{10} and PM\textsubscript{2.5} levels in the north of Tehran, which were found to be significantly higher than the national average on a per m\textsuperscript{2} basis. The excessive emission of pollutants and acidifying compounds such as nitrous oxides and sulphuric acid into the atmosphere favours conditions for acid rain in the form of fog, snow and rain (Uchiyama et al., 2017; Zheng and Yu Hong, 1994). Acid rain is defined as a rain characterized by pH values lower than 5.6 (Mirhosseini et al., 2009; Purohit, and Kakrani, 2002; Neill, 1993; Welburn, 1990). Acid rain adversely affects ecological and environmental processes (Wang et al., 2018; Wei et al., 2017). Over the last 50 years, several researchers have highlighted the negative environmental impacts of acid rain, including destruction of buildings and tools (Yokom and Beer, 1983), forest and geological formation degradation (Ulrich, 1980, Driscoll et al., 2001), and adverse effects on crops and soil fertility (Ferenbaugh, 1976; Pell et al., 1987; Irving, 1987).

Evidence is gathering that this acid rain is increasing close to highly populated and industrial areas in Iran. For example, Moarref et al. (2011) recorded pH values of about 2.4 by evaluating the chemical composition of rain in Ahwaz city. Mirhosseini et al. (2009) investigated the occurrence of acid rain in Sarcheshmeh area, located in the Kerman province, and determined that that occurrence is an inevitable phenomenon close to highly industrial and populated regions. However, these authors remarked that there is a lack of quantification of the effects of acid rain on soil processes such as water soil erosion (Mirhosseini et al., 2009; Moarref et al., 2011). As acid rain impairs soil fertility and vegetation development, the areas with bare soil surfaces will increase (Driscoll et al., 2001). Lack of vegetation is one of the most important driving factors of soil erosion processes in watersheds and hillslopes (Cerdà et al., 2018; Saleh et al., 2017; Parsakhoo et
Water soil erosion is initiated by the collision of rain drops with the soil surface (Ellison, 1944; Ellison, 1947; Free 1952). Rain drops can separate and move soil particles and aggregates (Fernández-Raga et al., 2017; Marzen et al., 2015).

The splash effect on soils is the first stage in the erosion process, which is the result of bombardment of the soil surface by rain droplets (Qinjuan et al., 2008, Wuddivira et al., 2009). The splash effect, by crushing soil particles and reducing their diameter, leads to a reduction of soil particle resistance against transport, and also a decrease in water penetration in soil surface layers, which in turn can increase runoff, erosion and sediment transport (Barry et al., 2010; Sadeghi et al., 2017).

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

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

The main aim of this study was to assess the impact of acidic rain at different pH levels and intensities on initial soil erosion processes under distinct soil conditions using a small portable
rainfall simulator. The experiments were conducted under laboratory conditions to avoid any external factors such as wind, soil property changes or inclinations. We hypothesize that this first approach will allow us to show if acidic rain *per se* directly affects initial soil erosion processes, as a basis for future research under natural conditions and in different environments.

2. Materials and methods

2.1. Soil sampling

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

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

The rainfall simulator was mounted on a metal A-frame structure. The height can range from 2 to 2.7 m depending on the purpose. The telescopic legs allow the height of the nozzles to be changed, which regulates the rainfall intensity and kinetic energy. The rainfall simulator can be used on rugged terrain as the telescopic legs allow levelling at any slope angle from 0 to 45°. Simulated rainfall is produced with two movable Veejet 80100 nozzles (Pall et al., 1983; Blanquies et al., 2003; Chouksey et al., 2017) with a diameter of 4.5 mm. Each nozzle is installed in a metal deposit to collect and reuse the excess of rainfall (that is not sprayed on the plot), and then returned to the pumping system. The rainfall simulator was used in the laboratory of the Sari Agricultural Sciences and Natural Resources University. The distilled water is pumped to the nozzles by means of a flexible hose with 15-mm diameter connected to an electric pump. The water pressure is monitored by a barometer installed in the transfer hose that allows the pressure to be regulated between 0 and 160 KPa (Fig. 1). A control board was designed with a programming capability of ten precipitation programs, to perform experiments with different rainfall characteristics. The control board can be used to set the velocity fluctuation nozzles, the oscillation angle of nozzles from 0° to 60°, and the duration of each rainfall event from 1 min to 1 hour. The plot size is 0.5 * 1 m² and 3 splash cups are located in the plot (figure 2). Splash cups were adapted from Morgan’s original design (Morgan, 1978).
Soil splash rates in eleven different treatments of rainfall acidity with dry and saturated soils, at three intensities of 40, 60 and 80 mm h\(^{-1}\), based on the average rainfall in three replicates, were recorded (Table 1). A total of 99 samples were tested.

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

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

2.3. Experimental procedure

Soil samples were located in the splash cups under dry and saturated conditions, using firstly non-acidic water and then acidic water with pH values of 3.75, 4.25 and 5.25. After 10 minutes of simulated rainfall, the splashed soil particles inside the splash cups after each treatment and intensity were separated in different containers at the end of the experiments. The yielded particles
were air-dried for 24 hours (Kavian et al., 2014). After the extra water was drained, the remaining sediment was transferred into suitable containers of specified weight and were dried in an oven at 105°C for 24 hours, then weighted using a scale (Sutherland & Ziegler, 1998).

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

\[ S = \frac{D_{t2} - D_{t1}}{(t_2 - t_1)A} \]  
(Eq. 1)

\[ S = \text{splash rate during the specified rainfall (gm}^2\text{minute)} \]
\[ D_{t1} = \text{Soil weight before splash experiment (g)} \]
\[ D_{t2} = \text{Soil weight after splash test (g)} \]
\[ \Delta t = (t_2 - t_1) \text{rainfall duration (min)} \]
\[ A = \text{area of splash cup (m}^2\text{)} \]

2.4. Statistical Analysis

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

3. RESULTS AND DISCUSSION
3.1 Comparison of drop splash effects on soil loss under dry conditions

Table 2 shows the results of one-way ANOVA of the different treatments at intensities of 40, 60 and 80 mm h\(^{-1}\) for the dry soil. Significant differences for the different treatments for an intensity of 40 mm h\(^{-1}\) at a confidence interval of 95% and an intensity of 60 mm h\(^{-1}\) at a confidence level of 99% are obtained. However, no significant differences are found for splash results with an intensity of 80 mm h\(^{-1}\). Considering the significant difference between the treatments at 40 mm h\(^{-1}\), Duncan's multiple range test was performed to show the difference among treatments. Figure 3 shows total initial soil erosion results and Duncan's multiple range test for control plot and acidic rain simulation under dry soil conditions. The highest particle loss due to splash effect in all degrees of rainfall intensities are observed with the most acidic rain (pH= 3.75), reaching maximum values of 16 g m\(^{-2}\) min\(^{-1}\). On the contrary, the lowest particle losses are obtained in the control plot, where non-acidic rain is used with values ranging from 3.8 to 8.1 g m\(^{-2}\) min\(^{-1}\). The results show for 40 mm h\(^{-1}\) that the AR1DS treatment is different (a) from the other three treatments, classified in group b. For a rainfall intensity of 60 mm h\(^{-1}\), the treatments are classified into three groups. The most acidic rain (AR1DS treatment) is in the first group (a) again. AR2DS and AR3DS appear in the same group (b) giving similar particle loss results, and the control plot (NARDS) is in group c. Finally, for 80 mm h\(^{-1}\) of rainfall intensity, the highest and the lowest values for splash were observed in the most acidic rain (AR1DS). The results are very similar with no significant difference among treatments at this high rainfall intensity, which even without acid rainfall, is able to contribute in bare soils to a high soil particle loss values (Beguería et al., 2015; Eldridge and Greene, 1994). Therefore, under dry conditions, we demonstrated that a higher concentration of acid rain is able to increase soil erosion rates. The low rates observed for the control plot and the lowest acidic rain (pH<5.25) could be due to the presence of calcium carbonate in our soil samples.
(25.3%), which was functioning as a stabilizing factor for aggregate stability (Bakhshipour et al., 2016). Therefore, parent material such as limestones or marls can act as a driving factor if carbonates are correctly transferred to soil horizons, as Cerdà (2002) showed in Mediterranean areas. However, when acid rain occurs, the lime present into the soils is neutralized; subsequently, soil particles can be more easily separated (Gratchev and Towhata, 2016).

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

Table 2 also shows the results of the ANOVA test for soils under saturated conditions for the intensities of 40, 60 and 80 mm h\(^{-1}\). For 40 mm h\(^{-1}\), there were no significant differences, despite the increasing trend in soil particle loss values of acid rainfall compared to non-acidic rain, though an increasing trend with more acidic rainfall was recorded. A significant difference in soil particle loss is found for treatments with rainfall intensities of 60 and 80 mm h\(^{-1}\).

Average values of total soil particle losses are shown in figure 4. Under saturated soil conditions, the smallest amount of soil particle loss was registered in the control plot, as also seen under dry soil conditions. The highest peaks of soil loss were obtained for the most acidic rains (pH= 3.75 and pH= 4.25), reaching maximum average values of 40 g m\(^{-2}\) min\(^{-1}\). However, for the Duncan test, the differences are not significant at 40 mm h\(^{-1}\), and only for 60 mm h\(^{-1}\) the most acidic rainfall shows a significant difference (AR3SS3). However, for 80 mm h\(^{-1}\), a significant difference was clearly seen among treatments, with the AR1SS1 treatment showing only slightly lower levels of soil particle loss than the AR2SS2 treatment. These findings confirm the importance of previous soil moisture in the plot. As other authors have found, saturated soils respond according to a Hortonian model, where runoff is able to activate soil loss when soil is saturated (Gabarrón-
Therefore, it is very important to pay attention to soil water content prior to performing the rainfall experiments, because this can impact loss rates (Hébrard et al., 2006; Wei et al., 2007). Kanga (1999) argued that when rainfall droplets hit the ground the soil particles are disintegrated, and then, due to the water content and their reduced adhesion force, particles are returned from the surface by droplets and soil particle loss increases. Our results confirm the contention that in arid and semiarid areas, soil water content is able to determine initial soil erosion rates, and an increase in acidification of the rainfall may also enhance these rates. This is an important finding which will affect many regions, particularly for calcareous soils in Iran. The rapid growth of urbanization and the increase in population close to large and industrial Iranian cities are increasing emissions of pollutants to the air (Fanni, 2006; Hafeznia et al., 2017; Yousefi et al., 2017), therefore, rapid solutions need to be found in order to stop this confirmed problem. As other researchers in Iran have found, there are other driving factors that also enhance soil erosion such as wind, bare soils and extreme rainfall events (Samani et al., 2016). This new factor, acidic rain, needs to be added to the list of potential drivers of soil erosion (Tarolli, 2016). Therefore, solutions or regulations to limit air pollution should be implemented in order to minimize, or stop, this confirmed land degradation process (Hansen et al., 2013; Smith et al., 2008).

3.3. Evaluation of the effects of non-acidic rainfall on acidified saturated soil at different rainfall intensities

The negative effects of acidic rain in calcareous soils are clear; however, in order to observe an inverse effect, the influence of non-acidic rain on acidic soils, we also conducted rainfall simulations on saturated soils at three different pH values, 3.75, 4.25, 5.25, with three different
rainfall intensities (40, 60 and 80 mm h\(^{-1}\)) but with non-acidic rain. Table 2 shows the results of one-way ANOVA for each treatment. There is no significant difference between the mean splash rate of NARSS, NARSS1, NARSS2, NARSS3 at intensities of 40 and 60 mmh\(^{-1}\), but there is for 80 mmh\(^{-1}\) at the 99% confidence level. Figure 5 shows the comparison among each treatment at three rainfall intensities (40, 60 and 80 mmh\(^{-1}\)) and pH levels using the Duncan's test at confidence interval of 95%. For a rainfall intensity of 40 mm h\(^{-1}\) and 60 mm h\(^{-1}\), there were no significant differences among treatments, registering the highest soil particle loss (17.1 g m\(^{-2}\) min\(^{-1}\)) in the control plot and the lowest one (10 g m\(^{-2}\) min\(^{-1}\)) for pH values of 5.25. With a rainfall intensity of 80 mmh\(^{-1}\), the control plot (NARSS) shows significant differences from the other treatments. The lowest soil particle loss was found for the pH value of 5.25 (18.7 g m\(^{-2}\) min\(^{-1}\)) with maximum rates in the control plot (32.9 g m\(^{-2}\) min\(^{-1}\)). This study shows the impact of soil saturation on increasing soil particle loss.

According to the comparison of means, the results showed that the lowest splash rate was attributed to the NARDS treatment. When comparing the average splash rate at the intensities of 40, 60 and 80 mmh\(^{-1}\), it was observed that splash rate increases with increasing intensity, confirming the findings of other studies (Gholami et al., 2016; Sadeghi et al., 2015); this means that with increasing rainfall intensity, the erosion rate increases, which can be due to the fact that with increasing rainfall intensity, the number of droplets that can hit the soil increases, and raindrops can increase in size and as a result their mass and falling velocity increase, which leads to more kinetic energy (Mohammadi and Kavian., 2015). The energy of the raindrops is a major factor in the disintegration of soil aggregates (Valettea et al., 2006; Barry et al., 2010; Brodowski et al., 2013), which can have a great influence on the separation of soil aggregates and results in splash erosion (Liu et al., 2016).
Also, changes in soil organic matter under short term acidic rainfall are plausible. As we observed in table 3, on dry and saturated soils, organic matter decreased after acidic rain from 1.24% to 0.97% and from 1.28% to 0.99%, respectively. Thus, further research is needed to investigate the loss of soil we are registering because of the dissolution of organic matter, but now over long-term periods.

### 3.4. Challenges and further investigations

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

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

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

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