

Biosolids Application in the Chihuahuan Desert: Effects on Runoff Water Quality

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ABSTRACT

Surface-applied biosolids, the option most often used on rangelands, can increase the concentration of macronutrients and trace elements in the runoff water and can potentially produce eutrophication or contamination of surface waters. In this study, the effects of postapplication age of biosolids (18, 12, 6, and 0.5 mo) and rate of application (0, 7, 18, 34, and 90 Mg ha⁻¹) on the quality of runoff water from shrubland and grassland soils were assessed. Between July and October 1996 simulated rainfall was applied to 0.50-m² plots for 30 min at a rate of 160 mm h⁻¹. All of the runoff water was collected. The concentration of NH₄⁺-N, NO₃⁻-N, PO₄³⁻-P, total dissolved phosphorus (TDP), Cu, and Mn in the runoff water increased with rate of biosolids application and decreased with time of postapplication on the two soils. The highest PO₄³⁻-P and NH₄⁺-N concentrations, 4.96 and 97 mg L⁻¹, respectively, were recorded in the grassland soil treated with 90 Mg ha⁻¹ of biosolids 0.5 mo postapplication. For the same soil, rate, and postapplication age of biosolids, Cu exceeded the upper limit (0.50 mg L⁻¹) in drinking water for livestock. Ammonium N and PO₄³⁻-P should be the main compounds considered when surface-applying biosolids. Ammonium N at concentrations found in all biosolids-treated plots may affect the quality of livestock drinking water by causing taste and smell problems. Orthophosphate can contribute to eutrophication if the runoff from biosolids-treated areas enter surface waters.

BIOSOLIDS, a by-product of municipal wastewater treatment plants, have been used on agricultural lands for a long time as an amendment to add plant macro- and micronutrients and organic matter to the soil. According to the USEPA (1993), land application accounted for 33.3% (1.79 million dry metric tons) of the municipal sludge generated nationwide in 1989. More recently, biosolids have been applied to native rangelands.

While biosolids applied to agricultural lands are generally incorporated into the soil, they have been surface-applied to rangelands to avoid soil disturbance and destruction of vegetation. Restrictions in biosolids disposal (e.g., ocean disposal banned in 1992) and an increasing social awareness concerning the need to recycle organic wastes to increase productivity and to achieve environmental sustainability make land application a viable alternative.

In arid rangelands, water tables are generally deep and the probabilities of leached nutrients or toxic elements reaching ground water are low. Arid land soils are generally calcareous throughout and have a high pH in the upper soil horizons favoring the precipitation of most heavy metals (Fuller, 1990). These factors decrease the risk of ground water contamination and the

risk of heavy metals entering the food chain via plant uptake.

Biosolids have a high content of plant macro- (N, P, and K) and micronutrients (Cu, Mn, Fe, and Zn) that may beneficially affect the low-fertility soils typical of desert ecosystems. The high organic matter content of biosolids may also improve the hydrologic properties of desert soils. A one-time biosolids application to degraded rangelands in New Mexico increased soil organic matter, N, biomass production, and soil infiltration capacity, and decreased erosion (Fresquez et al., 1990; Aguilar and Loftin, 1991). Surface application of biosolids to Chihuahuan desert grasslands in western Texas have also increased biomass production (Jurado-Guerra, 1996; Benton and Wester, 1998) and soil infiltrability and decreased erosion (Moffet, 1997).

In addition to adding plant nutrients, biosolids can also add potentially toxic elements such as Pb, Ni, and Cd. Acceptable biosolids management options must minimize negative environmental effects including degradation of the soil's capacity to sustain plant growth and discharge of nutrients, especially N and P, and other potentially toxic elements to surface and ground water. Both nutrients and toxic chemicals can be transported off-site by surface runoff. Most of the loss in runoff occurs from a few high-intensity rainstorms, and the concentrations of chemicals in runoff can be high during those events (Hubbard et al., 1982). Thus, surface runoff may transport bioavailable P that can accelerate surface water eutrophication (Sharpley and Menzel, 1987). Although both N and P stimulate eutrophication, P is generally the nutrient most influential in algae growth (Sharpley et al., 1994). Runoff water from arid rangelands may, in some cases, be collected in ponds for livestock or wildlife consumption. Water quality criteria for these uses have been established (Environmental Studies Board, 1973).

Several investigators have studied the effects of biosolids or wastewater application on runoff water quality from agricultural lands (Dunigan and Dick, 1980; Duncomb et al., 1982; Mostaghimi et al., 1992; Bruggeman and Mostaghimi, 1993). Surface-applied biosolids, the option most often used on rangelands, resulted in higher loading of N and P runoff as compared with areas in which the biosolids were incorporated into the soil (Dunigan and Dick, 1980; Bruggeman and Mostaghimi, 1993). According to Sharpley (1985), the first step in the movement of dissolved P in runoff is desorption, dissolution, and extraction of P from soil, crop residue, or manure. These processes occur as rainfall interacts with a thin layer (1–2.5 cm) of surface soil before leaving a field as runoff. The direct interaction of rainfall with surface-applied biosolids may explain the higher loading

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of P as well as other elements or compounds in runoff. Dowdy et al. (1994) found that where biosolids were applied annually by injection to an area cropped continuously with corn, Cd, Ni, and Pb concentrations in the runoff water were not affected by 10 yr of biosolids application. However, biosolids application increased Cu and Zn concentrations in snowmelt runoff (Dowdy et al., 1994).

Few studies dealing with the effect of biosolids application on runoff water quality have been conducted on rangelands (Aguilar and Loftin, 1991; Harris-Pierce et al., 1995). In a one-time application experiment with surface-applied biosolids, Aguilar and Loftin (1991) found that Cd, Cu, and NO_3^- -N concentrations in the runoff water produced either by natural or simulated rainfall were below established New Mexico livestock and wildlife watering standards. In a similar experiment, with a one-time surface application of 22 and 41 Mg ha^{-1} of biosolids to a semiarid grassland, Harris-Pierce et al. (1995) found that organic N and NH_4^+ -N, P, Na, K, Cu, Mo, and Ni concentrations in runoff water increased with increasing biosolids application rate compared with a control without biosolids application.

In commercial operations, biosolids are usually applied at different rates and at intervals of 1 to 3 yr in the same area. After a certain period on the ground, surface-applied biosolids undergo changes in composition that may attenuate the nutrient release and their effect on the quality of the runoff water produced. Concerning runoff from manured soils, Kirchmann (1994) concluded that the closer the rainfall event was to the date of application, the higher the nutrient concentrations were in the runoff water. We could expect that this generalization applies to surface-applied biosolids; however, there are no reported data on the effect of time after biosolids application on the quality of runoff water from western U.S. rangelands.

Therefore, the objective of this study was to determine the effect of surface-applied biosolids at different rates and dates on the runoff water quality from two Chihuahuan desert range sites. The variables tested for significant effects on the runoff water quality were (i) postapplication age of biosolids and (ii) biosolids application rate.

The hypothesis formulated in relation to the effect of postapplication age and rate of biosolids application on runoff water quality was that the electrical conductivity (EC), NO_3^- -N, NH_4^+ -N, PO_4^{3-} -P, and selected metals and metalloids would not increase as rate of biosolids application increased and time of postapplication decreased.

MATERIALS AND METHODS

Study Area

The study was conducted on the Sierra Blanca Ranch, 9 km north of Sierra Blanca in western Texas (Hudspeth County). The climate of the region is semiarid and warm. The average annual precipitation is 310 mm with about 65% falling between July and September. The mean annual temperature is 18°C (National Oceanic and Atmospheric Administration, 1994).

The physiography of the study area is typical of the northern Chihuahuan Desert: a large gently sloping to nearly level bol-

son, bounded by igneous hills on the west and a limestone escarpment on the north and east. Site elevation is approximately 1350 m. The bolson is drained by an ephemeral stream. Two range sites were selected for this study: a midsection of an alluvial fan where a Chilicotal soil occurs, and a distal section (toeslope position) of the alluvial fan where a Stellar soil dominates (Allen et al., 1993). The range site where the Stellar soil dominates is adjacent to the ephemeral stream and may occasionally receive runoff from the upper land units.

The USDA Natural Resources Conservation Service has not published a soil survey for Hudspeth County. The Chilicotal soil as mapped in the study area is a taxadjunct to the Chilicotal series and has been tentatively classified as a fine-loamy, mixed, superactive, thermic Ustic Calciargid (Casby-Horton, 1997). The A horizon (0–10 cm) of this soil has a pH of 8.1 and a CEC of 16 $\text{cmol}_c \text{kg}^{-1}$. This soil is calcareous throughout and the calcic horizon is developed at the base of the profile (140 to >190 cm) (Casby-Horton, 1997). The slope is 3 to 6%. Rills up to 1 m deep drain this site. Mound interspace areas are covered with a desert pavement that lies on top of a weak crust.

The Stellar soil as mapped in the study area is a taxadjunct to the Stellar series and has been tentatively classified as a fine, mixed, superactive, thermic Vertic Paleargid (Casby-Horton, 1997). The A horizon (0–7 cm) of this soil has a pH of 7.8 and a CEC of 21 $\text{cmol}_c \text{kg}^{-1}$. The profile of the Stellar soil is calcareous throughout with effervescence increasing with depth. Its slope is <1%. Bare, eroded areas ranging from 1 to 10 m^2 with a thick and strong crust (Av horizon 2.5 cm) that cracks when dry are common in the Stellar soil.

Vegetation of the midsection of the alluvial fan is a shrubland dominated by creosotebush [*Larrea tridentata* (Sess & Moc. ex DC) Coville] and is part of the gravelly range site in fair range condition (Wester and Benton, 1993) that occurs on the Chilicotal soil. Vegetation is highly aggregated in patches generally associated with mounds 20 to 30 cm in height and 1 to 3 m in diameter. Fluffgrass [*Erioneuron pulchellum* (Kunth) Tateoka], black grama [*Bouteloua eriopoda* (Torr.) Torr.], and sand dropseed [*Sporobolus cryptandrus* (Torr.) Gray] are the most abundant grasses on the range site. Ground cover characteristics of this range site were 3% basal vegetation cover, 26% gravel, 14% litter, and 3% cryptogamic cover. Bare ground comprised the other 54%. Degradation processes have included loss of grass cover and an increase in creosotebush and bare ground. Evidence of soil erosion includes a dense network of rills, mounds associated with shrubs, and a desert pavement in the intermound areas.

Vegetation of the distal section of the alluvial fan is a grassland dominated by tobosagrass [*Hilaria mutica* (Buckl.) Benth.] and is part of a loamy range site (Wester and Benton, 1993) that occurs on the Stellar soil. Codominant grasses are alkali sacaton [*Sporobolus airoides* (Torr.) Torr.] and blue grama [*Bouteloua gracilis* (Kunth) Lag. ex Griffiths, nom. illeg.]. Mesquite (*Prosopis glandulosa* Torr.) and lotebush [*Ziziphus obtusifolia* (Hook. ex Torr. & A. Gray) A. Gray] are widely scattered in the grassland. Ground cover characteristics of this site were 5% basal vegetation cover, 5% gravel, 6% cryptogamic, 24% litter, and 60% bare ground. This site was in mid to fair range condition (Wester and Benton, 1993). Degradation processes include an increase in bare ground and an increase in tobosagrass dominance. Evidence of erosion includes the formation of crusted bare patches where the A horizon has been eroded.

Experimental Procedures

In January and July 1995 and 1996 (18, 12, 6, and 0.5 mo before the initiation of the simulated rainfall application), biosolids were topically applied to 100 experimental units

(0.50-m² plots) on each range site. Each of these plots was located in bare areas. Rates of biosolids applied were equivalent to (dry weight basis) 0, 7, 18, 34, and 90 Mg ha⁻¹. The 18 Mg ha⁻¹ is the agronomic rate established by the Texas Natural Resources Conservation Commission (TNRCC), while the 90 Mg ha⁻¹ rate was used for experimental purposes only and is not considered to be a viable recommended rate.

Before each biosolids application, five samples of biosolids were collected for chemical analyses. In July 1996, before the simulated rainfall experiments were begun, biosolids samples for chemical analyses were also collected from the 90 Mg ha⁻¹ plots treated in 1995 and in January 1996. Prior to simulated rainfall application, a painted sheet metal frame (70 × 58 cm, 0.40 m²) was dug into the treated area to channel the runoff generated by the simulated rainfall on the experimental plot so that it could be collected. Runoff leaving the lower border of the plot was collected with a u-shaped collector provided with a cover to protect it from the rainfall.

Between July and October 1996, one simulated rainfall was applied to each plot with a portable single-nozzle rainfall simulator (Moffet, 1997) at a rate of 160 mm h⁻¹ and a duration of 30 min. In the study area, a rainfall with this intensity and duration has an expected frequency of less than once every 100 yr (U.S. Department of Commerce Weather Bureau, 1957).

Runoff was collected and weighed at 5-min intervals during each simulated rainfall event. All of the runoff was stored in 30-L plastic containers. Immediately after a simulated rainfall application, a 0.5-L sample of runoff water for chemical analysis was collected in acid-washed plastic bottles and filtered through a 0.45-μm membrane filter. A 100-mL sample for metal analyses was separated out and maintained at a pH <2 with concentrated nitric acid (USEPA, 1983). Dissolved Al, As, Ba, Be, Cd, Cu, Cr, Fe, Mn, Mo, Pb, Ni, P, Se, and Zn were analyzed by inductively coupled plasma emission spectroscopy (ICP). Nitrate N and orthophosphate ("reactive P") were determined colorimetrically by Cd reduction to nitrite (Greiss-Ilosvay method) (Keeney and Nelson, 1982) and the colorimetric method of Murphy and Riley (1962), respectively. Ammonium N was determined using an NH₃ ion-selective electrode. The EC, an estimate of dissolved solids in runoff water, was measured with a conductivity meter, and the pH was measured using an ion-selective electrode.

Statistical Analyses

The experimental design for this study was a split-split plot arrangement of a completely randomized design (Table 1) (Kirk, 1982). Soil was the main plot factor. Postapplication age of biosolids (four ages) was the subplot factor, and rate of biosolids application (five rates) was the sub-subplot factor. One hundred plots were randomly located (20 groups of five plots) on each range site. Postapplication age of biosolids was randomly assigned to the 20 groups of five plots; rate was

then randomly assigned to each plot in each group. Each age × rate combination was replicated five times. There were 20 treatment combinations on each range site.

Data normality was tested by means of the Shapiro-Wilk's test (SPSS, 1997). When data were not normally distributed, the Box and Cox (1964) diagnostic procedure was used to select the most appropriate transformation. Some variables (EC and NH₄⁺-N) were log transformed; Mn and Cu were transformed by square root. When an element or compound was below the detection limit of the analytical procedure used, the detection limit divided by two was used. If an element or compound was below the detection limit in all the plots of a particular treatment rate for the four postapplication ages (i.e., the control), the treatment rate was not considered in the analysis of variance (see Steel and Torrie, 1980, p. 170). Homoscedasticity in the main plot analysis was tested with Hartley's (1940) test. Mauchley's (1940) test was used to test for sphericity in the subplot analyses; when sphericity was not satisfied, Greenhouse-Geisser adjusted degrees of freedom were used in the *F* test. The data were statistically analyzed using the SPSS program (SPSS, 1997). Within this program, the GLM-Repeated Measure Analysis was used to test the differences among treatment means. Mean separation with the protected LSD test (*P* ≤ 0.05) was used to compare runoff quality variable means among treatments.

RESULTS AND DISCUSSION

Biosolids Composition

The average compositions of the fresh biosolids applied on the four application dates and the biosolids collected from the plots in July 1996 (date of the initiation of the simulated rainfall application) are shown in Table 2. Biosolids composition varied across the application dates. Total Kjeldahl nitrogen (TKN) was significantly higher in the biosolids applied in January 1995 than in the biosolids applied on the other dates. In general, metal concentrations were higher in the biosolids applied in July 1996 compared with the biosolids applied in January 1995. For example, Mn and Cd concentrations were three and two times higher in the July 1996 biosolids compared with the biosolids applied in January 1995.

The concentration of most metals and P in the biosolids applied in January and July 1995 (18 and 12 mo postapplication) increased by an order of 1.2 to 2 by the time we started to apply the rainfall (July 1996). On the contrary, the TKN (and probably organic matter) concentration decreased about 35% 1 yr postapplication. Biosolids organic matter decomposition and losses (volatilization, leaching, and runoff) of nitrogen compounds may account for these changes. Sosebee et al. (1993) reported that organic matter content of surface-applied biosolids decreased from 55 to 42% in a 6-mo period. The biosolids applied in July 1995 had the lowest organic matter and TKN, and the highest Fe content in the samples analyzed prior to the rainfall application.

Rainfall During the Sampling Period

Daily rainfall recorded during the period when simulated rainfalls were applied to treated plots (18 July to 17 Oct. 1996) is shown in Fig. 1. Two weeks after the last biosolids application (2 July 1996), a high-intensity rain (26.6 mm) occurred causing a temporary ponding

Table 1. Analysis of variance model for the different runoff water variables at Sierra Blanca, Texas.

Source of variation	df
Soil (S)	1
Error A	8
Post-application age (D)	3
S × D	3
Error B	24
Application rate (R)	4
S × R	4
D × R	12
S × D × R	12
Error C	128
Total	199

Table 2. Average biosolids chemical composition (dry matter basis) for the four application dates: fresh (sampled at the moment of application) and sampled in July 1996 ($N = 5$), Sierra Blanca, Texas. Mean comparisons are made, for each application date, between the variable values of the fresh biosolids and the variable values of the biosolids sampled in July 1996, and among the application dates, for the variable values of the fresh biosolids.

		Biosolids application dates						
		January 1995		July 1995		January 1996		July 1996
		Sampling dates						
Variable†	Units	Fresh	July 1996	Fresh	July 1996	Fresh	July 1996	Fresh
OM	%	nd‡	54.9	nd	50.4	nd	56.8	57.7
TKN	g kg ⁻¹	43.5A	30.2***	35.6B	22.8***	35.1B	26.1***	33.6B
P	g kg ⁻¹	16.9A	21.4***	14.9B	20.5***	19.4C	19.3(ns)	20.3C
S	g kg ⁻¹	nd	8.1	nd	8.1	nd	10.4	12.0
Ca	g kg ⁻¹	18.7A	23.8*	19.1A	24.7***	21.5B	22.7(ns)	25.5C
Mg	g kg ⁻¹	5.8A	6.8**	5.7A	6.0(ns)	7.6B	6.6*	6.6C
Na	g kg ⁻¹	nd	0.6	nd	0.6	nd	0.7	0.9
K	g kg ⁻¹	0.8A	1.5***	0.9A	1.7***	1.7B	1.8(ns)	1.9B
Fe	g kg ⁻¹	32.7A	27.3**	43.8B	41.7(ns)	28.2A	33.4(ns)	28.7A
Al	g kg ⁻¹	7.1A	11.1***	8.1B	13.0***	10.4C	10.5(ns)	11.7D
Cu	mg kg ⁻¹	890A	1132***	730B	802(ns)	909A	880(ns)	1103C
Mn	mg kg ⁻¹	395A	514**	663B	789**	1145C	1119(ns)	1248D
Zn	mg kg ⁻¹	882A	1125***	917A	1198**	1090B	1149*	1094B
Cd	mg kg ⁻¹	2.48A	4.8***	3.74B	5.8*	5.5C	5.6(ns)	4.74C
Ni	mg kg ⁻¹	22.7A	33.1***	25.3B	45.0***	41.0C	36.7**	41.0C
Pb	mg kg ⁻¹	176A	289***	233B	416***	374C	384(ns)	319D
EC	dS m ⁻¹	7.75AC	2.77***	8.86AB	2.19***	10.7B	3.18***	6.81C
pH		nd	6.69	nd	6.70	nd	6.51	6.33§

† OM, organic matter; TKN, total Kjeldahl nitrogen; EC, electrical conductivity.

‡ In a row, and for each application date, fresh and sampled on July 1996 biosolids are either not significant ($P \geq 0.05$) or *, **, and ***, significantly different at $P \leq 0.05$, ≤ 0.01 , and ≤ 0.001 , respectively. In a row, fresh biosolids with the same uppercase letters are not significantly different ($P \geq 0.05$). nd = no data.

§ Samples collected 45 d after biosolids application.

of water in the grassland area. This rain had been preceded by two 7-mm rains the previous 24 and 48 h. After simulated rainfall treatments were started on the shrubland soil, three rains >10 mm were recorded. The total amount of rainfall recorded during the simulated rainfall application period was 104 mm. The extent to which these rains could have affected the runoff water quality is discussed in the following paragraphs.

Runoff Water Quality: Electrical Conductivity and pH

The EC of the runoff water was significantly higher ($P \leq 0.05$) from the Stellar than from the Chilicotal soil. This difference may be in part explained by the lower leaching potential of the Stellar (low infiltration rate) than that of the Chilicotal soil (high infiltration rate). In the two soils the EC increased as rate of biosol-

ids application increased and as time of postapplication decreased (Fig. 2). Soluble salt content of the fresh biosolids decreased from 8 000 to 10 000 $\mu\text{S cm}^{-1}$ to 2000 to 3000 $\mu\text{S cm}^{-1}$ after 6 mo in the field. Thus, after a few months of laying on the soil surface, soluble salts released by biosolids decreased significantly. The EC of the runoff water from the plots treated in January 1995 with 90 Mg ha⁻¹ of biosolids, in the Chilicotal soil, was five times lower than the plots treated at the same rate in July 1996 (Fig. 2). Runoff water from the 90 Mg ha⁻¹ plots on the Stellar soil treated in July 1996 had the highest EC, 600 $\mu\text{S cm}^{-1}$. Total dissolved solids of 1000 mg L⁻¹ ($<1500 \mu\text{S cm}^{-1}$) is considered safe for both livestock and wildlife (Van der Leeden et al., 1990). The Environmental Studies Board (1973) considered water with up to 7000 mg L⁻¹ (11 000 $\mu\text{S cm}^{-1}$) as reasonably safe for cattle and sheep.

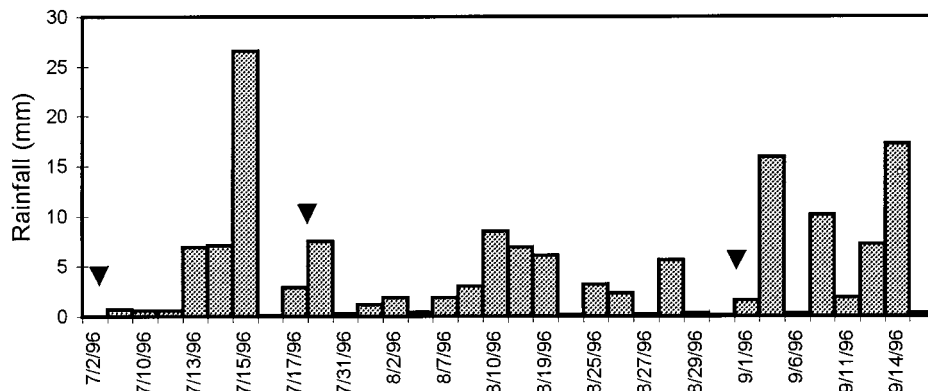


Fig. 1. Natural rainfall events and amount during the sampling period (18 July to 17 Oct. 1996), Sierra Blanca, Texas. Arrows correspond to biosolids application date (last application) and start of simulated rainfall on the Stellar and Chilicotal soils, respectively. Dates on the horizontal axis are listed in the month/day/year format.

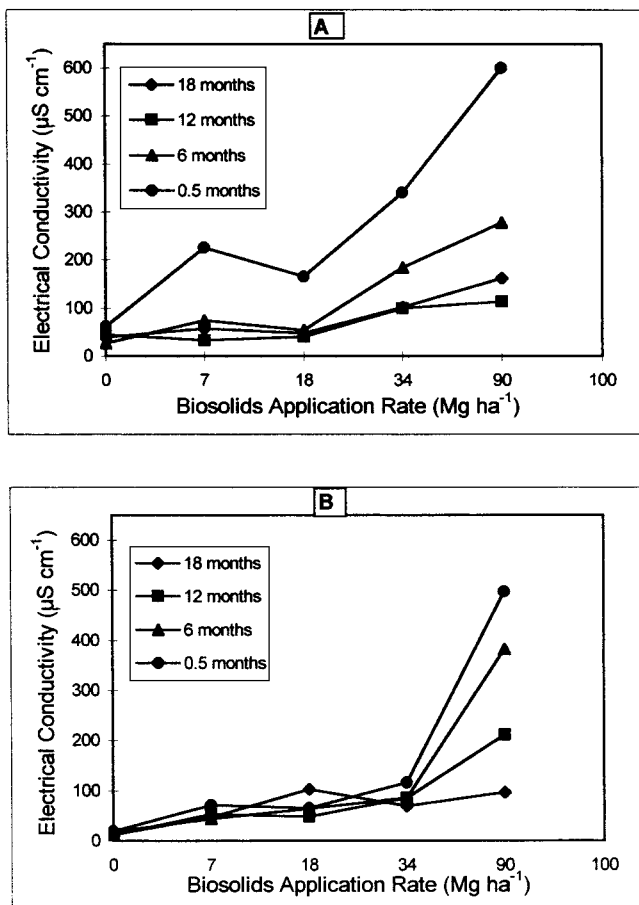


Fig. 2. Electrical conductivity (EC) of the runoff water of the Stellar (A) and Chilicotl (B) soils treated 18, 12, 6, and 0.5 mo before the simulated rainfall initiation with 0, 7, 18, 34, and 90 Mg ha⁻¹ of biosolids, Sierra Blanca, Texas.

The pH of runoff water was significantly lower ($P \leq 0.05$) from the Stellar than from the Chilicotl soil; for the two soils, pH decreased as biosolids application rate increased. Postapplication age of biosolids did not affect ($P \leq 0.05$) the pH. In the 90 Mg ha⁻¹ plots, the pH decreased between 0.32 to 0.43 units compared with the control plots for the Chilicotl soil, and from 0.57 to 1.06 units in the Stellar soil. The major decrease was recorded in the plots treated in July 1996 (Tables 3 and 4). However, the pH of runoff water was within the range tolerated for most aquatic biota (Novotny and Olen, 1994).

Nitrate

The NO₃⁻-N concentration of runoff water was above the detection limit (0.1 mg L⁻¹) in many of the treated plots in the Stellar soil, particularly following the last date of application. In the Chilicotl soil, NO₃⁻-N concentration was above the detection limit (0.1 mg L⁻¹) only in the plots treated with 18, 34, and 90 Mg ha⁻¹ of biosolids in July 1996, and in the 90 Mg ha⁻¹ rate for all biosolids postapplication ages (Tables 3 and 4). The NO₃⁻-N concentration was significantly higher ($P \leq 0.05$) from the Stellar than from the Chilicotl soil. The infiltration rate was lower in the Stellar than in the Chilicotl soil. Thus, in the Chilicotl soil the potential

for leaching was higher than in the Stellar soil. Nitrate concentration in the runoff water was significantly affected ($P \leq 0.05$) by rate and time after biosolids application. There was also a significant ($P \leq 0.05$) interaction between rate and time after biosolids application. Nitrate concentration in the runoff water of the two soils increased as rate of biosolids application increased (Tables 3 and 4). Although the lowest values were recorded in the plots treated in 1995, the effect of time after biosolids application was not consistent since the lowest concentrations were not found in the plots treated in January 1995, 18 mo before the simulated rainfall application, but in those plots treated in July 1995, 12 mo before the simulated rainfall. The biosolids applied in July 1995 had the lowest total N content at the time of simulated rainfall application (Table 2). In both soils, the highest concentrations of NO₃⁻-N were found in the 90 Mg ha⁻¹ plots treated in July 1996, with concentrations of 1.1 and 2.1 mg L⁻¹ recorded in the Chilicotl and Stellar plots, respectively. In the Chilicotl plots treated with less than 18 Mg ha⁻¹, NO₃⁻-N was below the detection limit for three of the four application dates (Table 4). From all of the plots, NO₃⁻-N concentrations in the runoff water were well below the maximum recommended level in drinking water for livestock (100 mg L⁻¹) as established by the Environmental Studies Board (1973).

It seems that the main path of movement of NO₃⁻-N in the soil is its downward movement with percolating water. It is possible that most inorganic N in biosolids remain as NH₄⁺-N, with the process of nitrification occurring once the NH₄⁺-N is leached out of the biosolids into the soil. Ryan et al. (1973), in an incubation experiment, found that nitrification slows down and NH₄⁺-N accumulates at high rates of biosolids application as compared with lower rates. Ammonium N accumulation in the biosolids may be easily removed by runoff water and, as with NO₃⁻-N, it might cause some risk of surface water contamination.

Ammonium

Ammonium N was the main form of inorganic N present in the runoff water from both the Stellar and Chilicotl soils. Soil, time after application of biosolids, and biosolids application rate significantly ($P \leq 0.05$) affected NH₄⁺-N concentration in the runoff water; the two- and three-way interactions were also significant.

The NH₄⁺-N concentration in the runoff water was higher in the Stellar than in the Chilicotl soil. Differences between soils were higher at the higher application rates (34 and 90 Mg ha⁻¹) and for the plots treated 0.5 mo before the simulated rainfall application was begun (July 1996). Part of the difference in NH₄⁺-N concentration between the two soils may be due to differences in the dates of simulated rainfall application. In the Stellar plots, simulated rainfall was applied between mid-July and the end of August; in the Chilicotl soil, simulated rainfall was applied between the beginning of September to mid-October.

Ammonium N increased from below the detection limit in most of the control plots to an average of 97.0 and 59.3 mg L⁻¹ in the 90 Mg ha⁻¹ plots treated in July

1996 in the Stellar and Chilicotal soils, respectively (Fig. 3). The relationship between $\text{NH}_4^+\text{-N}$ concentration in the runoff water and biosolids application rate in the Stellar plots treated in July 1996 was approximately equivalent to $1 \text{ mg } \text{NH}_4^+\text{-N } \text{L}^{-1}$ to $1 \text{ Mg biosolids } \text{ha}^{-1}$, for the 34 and $90 \text{ Mg } \text{ha}^{-1}$ application rates. Ammonium N content of fresh biosolids applied in 1997 on the Sierra Blanca Ranch, produced by the same wastewater treatment plants that produced the biosolids applied in 1995 and 1996, was approximately $7.5 \text{ g } \text{kg}^{-1}$.

In the plots treated in January and July 1996, the NH_4 concentration in runoff water significantly increased ($P \leq 0.05$) from both soils at each increasing rate of biosolids application. However, there were no significant ($P \geq 0.05$) differences between the control and the $7 \text{ Mg } \text{ha}^{-1}$ treatments in the two soils treated in January 1995. In the July 1995-treated plots of the Stellar soil, differences among the control and the 7 and $18 \text{ Mg } \text{ha}^{-1}$ treatments were not significantly ($P \geq 0.05$) different.

Eighteen months after biosolids were applied to plots in January 1995, the $\text{NH}_4^+\text{-N}$ concentration in runoff water from the $90 \text{ Mg } \text{ha}^{-1}$ plots decreased 2.6 times on the Chilicotal soil and 3.7 times on the Stellar soil with respect to the $\text{NH}_4^+\text{-N}$ concentrations found in the runoff water from more recently treated plots (July 1996). Differences in $\text{NH}_4^+\text{-N}$ were greater for the lower application rates (Fig. 3). As with $\text{NO}_3^-\text{-N}$, the lowest concentration of $\text{NH}_4^+\text{-N}$ for almost all of the rates was recorded in the plots treated in July 1995. Biosolids sampled in plots treated in July 1995 had the lowest N content (Table 2).

In the Meadow Spring Ranch study where biosolids were also surface-applied, Harris-Pierce et al. (1995) found a similar $\text{NH}_4^+\text{-N}$ concentration in the runoff water. In this study, in which simulated rainfall was applied 2 wk after the biosolids were applied, they found 51.3 and $76.0 \text{ mg } \text{L}^{-1}$ of $\text{NH}_4^+\text{-N}$ in the runoff water for the 22 and $41 \text{ Mg } \text{ha}^{-1}$ application rates, respectively. Part of this $\text{NH}_4^+\text{-N}$ may have been carried with the sediments. The $\text{NH}_4^+\text{-N}$ content of the biosolids at the time of application was $11 \text{ g } \text{kg}^{-1}$.

It is apparent that the potential for $\text{NH}_4^+\text{-N}$ losses in the runoff water from areas treated with surface-applied biosolids is high, especially when rainfall occurs soon after biosolids application. Mostaghimi et al. (1992) compared the water quality from conventional-tillage plots with biosolids incorporated into the soil and surface-applied biosolids. The $\text{NH}_4^+\text{-N}$ concentration from plots where biosolids were surface-applied averaged $25.8 \text{ mg } \text{L}^{-1}$, while in the plots where biosolids were incorporated, the $\text{NH}_4^+\text{-N}$ concentration was sixfold less than from surface-applied treatments, with an average of $3.8 \text{ mg } \text{L}^{-1}$. In both cases, biosolids containing $9.6 \text{ g } \text{NH}_4^+\text{-N } \text{kg}^{-1}$ had been applied at a rate of $11.5 \text{ Mg } \text{ha}^{-1}$.

Ammonium N is rapidly nitrified in well-aerated soils, provided soil air and water are adequate; in most soils this is near field capacity water content (Prasad and Power, 1997). Ammonium N in the runoff water is not toxic per se although it can give rise to NH_3 if the pH is alkaline. Ammonia gas can be produced that may adversely affect several species of fish. Concentrations as low as $0.02 \text{ mg } \text{NH}_4^+\text{-N } \text{L}^{-1}$ may increase the risk of

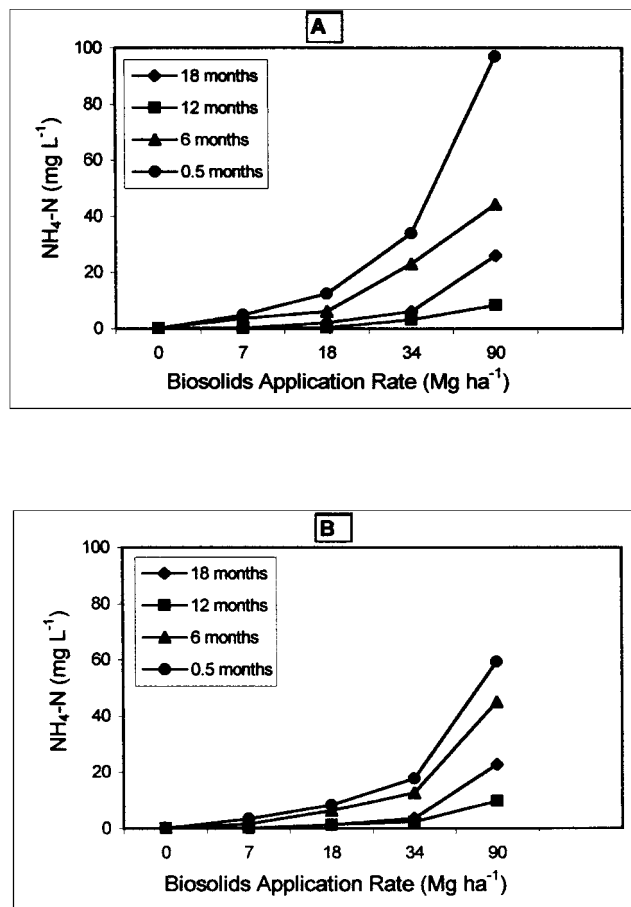


Fig. 3. Ammonium N in the runoff water of the Stellar (A) and Chilicotal (B) soils treated 18, 12, 6, and 0.5 mo before the simulated rainfall initiation with 0, 7, 18, 34, and $90 \text{ Mg } \text{ha}^{-1}$ of biosolids, Sierra Blanca, Texas.

fish asphyxia if converted to NH_3 (McNeely et al., 1980). Approximately 1% of NH_4 in aqueous solution at a pH 7.2 (20°C) occurs in the NH_3 form (Koelliker and Miner, 1973). Although there are no recommendations for $\text{NH}_4^+\text{-N}$ for livestock drinking water, ammonium at concentrations of $0.1 \text{ mg } \text{L}^{-1}$ may cause taste and smell problems (Boucher, 1985).

Orthophosphate

Orthophosphate concentrations in the runoff water of the biosolids-treated plots were higher ($P \leq 0.05$) in the Stellar than in the Chilicotal soil. Postapplication age and rate of biosolids application and the two-way interactions also had a significant effect on the concentration of $\text{PO}_4^{3-}\text{-P}$ in the runoff water.

For the plots treated with $90 \text{ Mg } \text{ha}^{-1}$ of biosolids in January and July 1995 and 1996, the $\text{PO}_4^{3-}\text{-P}$ concentrations in the runoff water of the Chilicotal soil were 24.8, 19.3, 6.4, and 16.5% lower than in the Stellar soil (Fig. 4). However, $\text{PO}_4^{3-}\text{-P}$ in runoff water was significantly different ($P \leq 0.05$) for the January 1995-treated plots.

Two factors might account for these differences. First, simulated rainfall was applied 1 wk to 3 mo later in the Chilicotal plots, so these plots were subjected to different environmental conditions than plots on the Stellar soil. Edwards and Daniel (1994) reported that

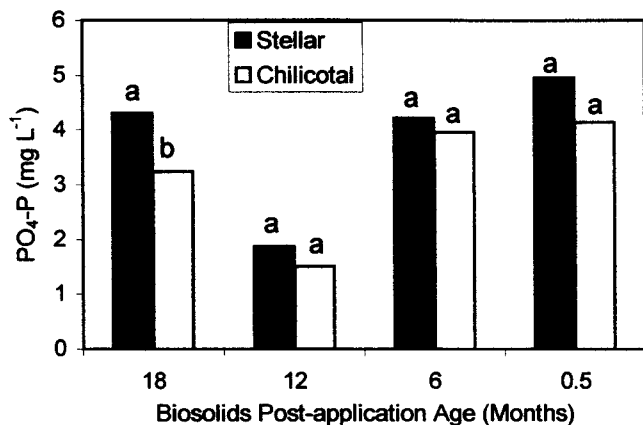


Fig. 4. Orthophosphate concentration in the runoff water of the Stellar and Chilicotal soils in the plot treated 18, 12, 6, and 0.5 mo before the simulated rainfall initiation with 90 Mg ha⁻¹ of biosolids, Sierra Blanca, Texas. Pairs of columns with the same lowercase letters were not significantly ($P \geq 0.05$) different.

$\text{PO}_4^{3-}\text{-P}$ concentration in runoff water from poultry litter-treated plots subjected to five successive 41-mm rainfalls decreased from $>11 \text{ mg L}^{-1}$ in the first runoff to control levels ($<3 \text{ mg L}^{-1}$) for the second to the fifth runoff collected 7, 14, 36, and 68 d after the first event. Second, the slope and the infiltrability of the two soils were different. In the Stellar soil, the average slope was 0.84% and in most cases it was concave, which increased the surface water retention capacity of the plots. In the Chilicotal soil, the average slope was 2.41% and was either straight or convex. The infiltration rate was lower in the Stellar than in the Chilicotal, thus ponded water was present from the beginning of the simulated rainfall application in the Stellar soil, and the layer of water ponded was deeper than in the Chilicotal soil. In these plots, the intimate contact between the slowly flowing layer of ponded water in the plots and the biosolids laying on the soil surface increased the opportunity for the desorption, dissolution, and removal of P from the biosolids by the runoff water and may explain the high $\text{PO}_4^{3-}\text{-P}$ concentration in runoff water flowing through surface-applied biosolids. In the Stellar soil, where the layer of water in the plot was thicker, a major solubilization might also have occurred. The 2- to 3-cm biosolids layer was composed of aggregates of different sizes and produced a high macroporosity in contrast with the crusted soil underneath. Heathman et al. (1995) and Ahuja et al. (1983) reported that concentration of Br in runoff water from soils with a 1- and 2.5-cm layer of 4.5- to 12.5-mm and 4- to 20-mm surface aggregates, respectively, increased almost 40 and 48 times compared with a control without a layer of aggregates. Large aggregates on the soil surface seem to increase the transfer of dissolved elements due to possibly deeper and greater turbulence in mixing rainwater and soil solution in large macropores.

In the Chilicotal soil, and for the highest rate of biosolids application (90 Mg ha⁻¹), there were no significant differences ($P \geq 0.05$) in $\text{PO}_4^{3-}\text{-P}$ concentration between the plots treated either in January 1995, January 1996, or July 1996 (3.24, 3.95, and 4.14 mg $\text{PO}_4^{3-}\text{-P L}^{-1}$, respectively). However, these treatments produced a signifi-

cantly ($P \leq 0.05$) higher $\text{PO}_4^{3-}\text{-P}$ concentration than the plots treated in July 1995 (1.51 mg L⁻¹) (Table 4). Although the total P content in the different biosolids batches was almost the same before the simulated rainfall application began, the Fe content of the biosolids applied in July 1995, as analyzed in July 1996, was almost 1% higher than in the other biosolids batches (Table 2). High Fe content in the biosolids may immobilize P (Soon et al., 1978). In the plots treated with 7, 18, and 34 Mg ha⁻¹, the $\text{PO}_4^{3-}\text{-P}$ concentration in the runoff water was significantly lower ($P \leq 0.05$) for the plots treated 18 and 12 mo before the simulated rainfall application than for the plots treated 6 and 0.5 mo before the simulated rainfall application. The effect of time after biosolids application on $\text{PO}_4^{3-}\text{-P}$ concentration is illustrated for the 90 Mg ha⁻¹ plots for the two soils (Fig. 4). As with $\text{NH}_4^+\text{-N}$, the plots treated in July 1995 released the least $\text{PO}_4^{3-}\text{-P}$. However, time after biosolids application had less effect on the $\text{PO}_4^{3-}\text{-P}$ than on the $\text{NH}_4^+\text{-N}$ concentration in the runoff water. Thus, while the concentration of $\text{PO}_4^{3-}\text{-P}$ in the runoff water from the 90 Mg ha⁻¹ treatment of the Stellar soil decreased 1.15 times in an 18-mo period (January 1995 vs. July 1996 plots), the concentration of $\text{NH}_4^+\text{-N}$ decreased 3.73 times for the same period.

The effect of rate of biosolids application on the $\text{PO}_4^{3-}\text{-P}$ concentration of the runoff water varied with postapplication age of biosolids. For the plots treated 18, 6, and 0.5 mo before the simulated rainfall application (January 1995, January 1996, and July 1996, respectively) on the Stellar soil, the $\text{PO}_4^{3-}\text{-P}$ concentration increased significantly ($P \leq 0.05$) at each biosolids application rate (Table 3). In the plots treated 12 mo before the simulated rainfall application (July 1995), although the $\text{PO}_4^{3-}\text{-P}$ in the plots treated with biosolids was higher than in the control, the difference between the 7 and 18 Mg ha⁻¹ treatments was not significant ($P \geq 0.05$). For the plots treated with 7 Mg ha⁻¹ of biosolids in the Stellar soil, the $\text{PO}_4^{3-}\text{-P}$ concentration in the runoff water was 5 to 12 times higher than in the control plots.

On the Chilicotal soil, the $\text{PO}_4^{3-}\text{-P}$ concentration in the runoff water of the 7 Mg ha⁻¹ plots was between two and four times higher than in the control plots (Table 4). Based on long-term evaluations of biosolids use over periods ranging from 9 to 23 yr, the Water Environment Research Foundation (1993) has recommended soil P levels be monitored where biosolids are used continuously over time, and that biosolids application rates be based on crop P levels rather than the N needs of the crop. Phosphorous removal rate for most crops has been summarized (Pierzynski and Logan, 1993).

The runoff water of the control plots had almost the same $\text{PO}_4^{3-}\text{-P}$ concentration for the two soils, except for the July 1996 plots in the Chilicotal soil where $\text{PO}_4^{3-}\text{-P}$ was highest. High litter cover on some of the plots of this treatment may account for this high value. For example, in two control plots where litter cover was exceptionally high ($>20\%$), the $\text{PO}_4^{3-}\text{-P}$ content of the runoff water was 0.25 and 0.53 mg L⁻¹. In both cases, a high proportion of the litter was washed off of the plots by the runoff water.

The $\text{PO}_4^{3-}\text{-P}$ concentration in the runoff water was above 0.03 mg L^{-1} in all of the treatments, a value considered a threshold for eutrophication in fresh water (Pierzynski et al., 1994). However, Sawyer and Engl (1947) indicated that 0.01 mg L^{-1} of inorganic P will enhance algae growth in streams and lakes.

Total Dissolved Phosphorus

Total dissolved P (TDP) concentrations ($\text{PO}_4^{3-}\text{-P}$ + organic P) in the control plots were essentially equal to the concentration of $\text{PO}_4^{3-}\text{-P}$. In the biosolids-treated plots, the differences increased as rate of biosolids application increased, with the highest differences occurring in the 90 Mg ha^{-1} plots where TDP was 53% higher (Chilicotal soil) than the $\text{PO}_4^{3-}\text{-P}$ (Table 4). The increasing amount of organic matter passing through the $0.45\text{-}\mu\text{m}$ membrane filter in the higher biosolids application rates may explain these differences. An exception was the plots treated in July 1995 in Stellar soil, where TDP was almost the same as $\text{PO}_4^{3-}\text{-P}$ (Table 3).

Trace Elements

Trace contaminants in public water supplies are usually arbitrarily defined as those for which drinking water standards are generally in the range of 1 mg L^{-1} or less (Sawyer et al., 1994). All trace elements in the runoff water from biosolids-treated plots were below this value, and most were below the method detection limits, or present in very low concentrations. Some metals and metalloids were below the method detection limit in all of the plots (i.e., As, Be, Cd, Pb, Cr, Ni, Co, Mo, and Zn), while some were below the detection limit in most of the biosolids-treated plots. Barium, for example, was above the detection limit (0.002 mg L^{-1}) in 17 out of the 20 control plots on the Chilicotal soil, with a mean concentration of 0.005 mg L^{-1} . However, it was below the detection limit in the plots treated with biosolids. This was probably due to interference in the matrix of the runoff water from the biosolids-treated plots. Strontium was above the detection limit in almost all of the blanks (Sierra Blanca tap water), but it was below the blank concentration in all the plots.

Copper

Copper concentration in the runoff water was significantly ($P \leq 0.05$) affected by soil, rate, and postapplication age of biosolids. The rate \times age interaction was also significant. Copper concentration in the runoff water was higher in the Stellar than in the Chilicotal soil (20 to 40% in the 90 Mg ha^{-1} plots). On the Stellar plots, simulated rainfall was applied 45 d earlier than on the Chilicotal plots. This difference in the time of simulated rainfall application may explain, in part, the difference in Cu concentration between the two soils. This is consistent with the results of Silveira and Sommers (1977), who found in a soil-biosolids incubation experiment that water-soluble Cu decreased from 0.8 to 0.1 mg L^{-1} in a period of 28 d. The thick layer of ponded water in the Stellar plots may have also increased the solubilization of Cu.

Copper concentration in the runoff water was below

the detection limit from all of the control plots. It increased as biosolids application rate increased, but it decreased with time of biosolids postapplication on both the Stellar and Chilicotal soils (Tables 3 and 4).

In the plots treated in 1996 on the Chilicotal soil, Cu significantly ($P \leq 0.05$) increased with each rate increase of biosolids application. From the Stellar soil, the Cu concentration of the runoff water from the 18 (recommended agronomic rate), 34, and 90 Mg ha^{-1} treatment rates of the January 1995, January 1996, and July 1996 treatment dates was significantly ($P \leq 0.05$) higher than the Cu concentration in the runoff from the control and the 7 Mg ha^{-1} treatments (Table 3).

There was also a significant interaction ($P \leq 0.05$) between rate and time after biosolids application in the resulting concentration of dissolved Cu. From the Chilicotal soil, postapplication age of biosolids affected Cu concentration in the runoff water at the four rates of biosolids application. For the highest rate of biosolids application, 90 Mg ha^{-1} , Cu concentration in the runoff water was significantly ($P \leq 0.05$) higher from the July 1996 (0.5 to 3 mo postapplication age) treated plots than from the other three postapplication ages. The lowest value for this treatment rate was recorded from the July 1995 (12 mo postapplication age) treated plots (Table 4). A similar age effect was recorded in the Stellar soil. The high concentration of Cu in the runoff water of the plots treated with 90 Mg ha^{-1} of biosolids in July 1996 probably resulted from stable complexes it forms with soluble organic materials present in biosolids. Copper soluble organic complex formation decreases as organic matter in the biosolids is decomposed. However, water-soluble forms of Cu in biosolids were calculated to be no greater than 0.3% of the total (Jenkins and Cooper, 1964), although Lagerwerff et al. (1976) extracted approximately 2% of Cu soluble in water from digested biosolids.

In both soils, the highest concentration of Cu was recorded in the 90 Mg ha^{-1} plots treated in July 1996 (0.5 mo postapplication age). In these plots the average concentration was 0.51 mg L^{-1} for the Stellar and 0.40 mg L^{-1} for the Chilicotal soil. Copper concentration in the July 1996-applied biosolids was 1103 mg kg^{-1} at the 90 Mg ha^{-1} rate of biosolids application, which is equivalent to 99 kg ha^{-1} of Cu applied. For the 90 Mg ha^{-1} application rate, equivalent to 13 times the commercial application rate, the lowest Cu concentrations in the runoff water were recorded from the July 1995-treated plots, and were 0.13 and 0.08 mg L^{-1} for the Stellar and Chilicotal soils, respectively. Differences in Cu content of the biosolids applied at different dates could explain the differences in Cu concentrations in the runoff water. For example, the Cu content in the fresh biosolids applied on July 1996 was 1.51 times higher than the Cu in the biosolids applied on July 1995 (1103 mg kg^{-1} vs. 730 mg kg^{-1}) (Table 2). However, the Cu concentration in the runoff water from the Stellar soil treated on July 1996 with 90 Mg ha^{-1} was 3.92 times higher than the Cu in the runoff water of the July 1995 plots treated with the same rate (0.51 mg L^{-1} vs. 0.13 mg L^{-1}). Similar results were found by Dowdy et al. (1991) in the Rosemount Watershed, Minnesota, where

liquid sludge was applied at 15 Mg ha⁻¹ (dry weight) for five consecutive years. The highest Cu concentration in the runoff water recorded in the spring of the fifth year was 0.092 mg L⁻¹. Additional sludge (5 Mg ha⁻¹) had been applied to frozen, snow-covered surfaces the previous winter. According to Dowdy et al. (1991), the extra application of sludge in conjunction with the climatic conditions would account for the higher Cu concentration in the spring runoff. Copper concentration in the sludge was 1030 mg kg⁻¹, with an application rate equivalent to 75 kg ha⁻¹. Harris-Pierce et al. (1995) found in plots treated with 22 and 41 Mg ha⁻¹ of biosolids that Cu concentration in the runoff water was 0.042 and 0.062 mg L⁻¹, respectively. The copper content of the biosolids applied in these experiments was 665 mg kg⁻¹, 60% of the Cu content of the biosolids that was applied in July 1996 in the Sierra Blanca study. To further place these Cu levels in perspective, the maximum recommended level in drinking water for livestock is 0.50 mg L⁻¹ (Environmental Studies Board, 1973).

Manganese

Manganese concentration in the runoff water was not affected ($P \geq 0.05$) by soil. It was significantly ($P \leq 0.05$) affected by biosolids application date and rate, and the soil \times date, soil \times rate, and date \times rate interactions. In the plots where biosolids had been applied 18 and 12 mo prior to the simulated rainfall application to the 7 Mg ha⁻¹ on the Stellar soil, the Mn concentrations in runoff water were below the detection limit (Table 3). However, Mn was detected in all the plots on the Chilicotal soil (Table 4), and in the plots treated in 1996 on the Stellar soil (Table 3). The concentration of Mn in natural waters is usually <1.0 mg L⁻¹, although there are some unusual waters in which the Mn contents are somewhat higher than 10 mg L⁻¹ (water with pH < 6) (Faust and Aly, 1981). Manganese in natural waters represents a nuisance type of constituent. Manganese usually enters the waters as dissolved Mn⁺⁺, which is slowly oxidized to black colloidal particles of Mn₂O₃. This precipitate imparts turbidity to water (Faust and Aly, 1981), but it is not a health risk. There is no recommendation level for Mn in drinking water for livestock.

The relatively high concentration of some elements (i.e., Cu and Mn) in the runoff water from biosolids-treated plots may in part be explained by the close contact between the applied water and the biosolids, since ponded water was present in almost the entire plot after 5 to 10 min of simulated rainfall application. This seems to be a fundamental difference in terms of runoff water quality between surface-applied biosolids and biosolids incorporated into the soil as used on agricultural lands.

CONCLUSIONS

The effect of postapplication age of surface-applied biosolids (18, 12, 6, and 0.5 to 3 mo) at five rates (0 to 90 Mg ha⁻¹) on runoff water quality in a shrub (Chilicotal taxadjunct soil) and in a grassland (Stellar taxadjunct soil) plant community of the Chihuahuan desert was studied. Most of the elements and compounds measured

were strongly affected by soil type, postapplication age of biosolids, rate, and the interaction between rate and postapplication age of biosolids. The EC of the runoff water increased with biosolids rate and decreased with time of biosolids postapplication; the pH followed an opposite trend. In general, NO₃⁻-N, NH₄⁺-N, PO₄³⁻-P, TDP, Cu, and Mn, concentrations in the runoff water increased with biosolids application rate and decreased as time of biosolids postapplication increased; these variables were higher in the Stellar than in the Chilicotal soil. The maximum NO₃⁻-N recorded was 2.1 mg L⁻¹, well below the maximum recommended level in drinking water for livestock (100 mg L⁻¹). Ammonium N was the main compound of inorganic nitrogen in the runoff water, and in the plots treated with 90 Mg ha⁻¹ of biosolids 0.5 mo before the simulated rainfall initiation it attained levels up to 100 mg L⁻¹. The PO₄³⁻-P was well above the background levels in all the treated plots. Postapplication age of biosolids had less effect on the concentration of PO₄³⁻-P than on the concentration of NH₄⁺-N. The lower concentrations of PO₄³⁻-P were found in the plots treated in July 1995, probably related to the higher content of Fe in the biosolids applied on that date. The highest Cu concentration in the runoff water was equivalent to the maximum recommended level in drinking water for livestock (0.50 mg L⁻¹). Except for Cu and Mn, all of the trace elements analyzed were below the method detection limits.

Phosphate P can contribute to the eutrophication of surface waters, should the runoff from biosolids treated areas enter a stream, while NH₄⁺-N may affect the quality of drinking water for livestock by changing the taste and smell of water collected in ponds.

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