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IMPACT OF POST-METHANATION DISTILLERY EFFLUENT IRRIGATION ON GROUNDWATER QUALITY

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Abstract. Molasses-based distilleries generate large quantities of effluent, which is used for irrigation in many countries including India. The effluent is rich in organic and inorganic ions, which may leach down and pollute the groundwater. An on-farm experiment was conducted to assess the impact of long-term irrigation with post-methanation distillery effluent (PMDE) on nitrate, sulphate, chloride, sodium, potassium, and magnesium contents in the groundwater of two sites in northwest India. Electrical conductivity (EC), pH, total dissolved solids (TDS), sodium adsorption ratio (SAR) and colour were also determined to assess the chemical load in the groundwater. Nitrate content in the groundwater samples ranged from 16.95 mg L^{-1} in the unamended fields to 59.81 mg L^{-1} in the PMDE-amended fields during the 2-year study (2001–2002). Concentrations of TDS in water samples from tubewell of the amended field was higher by 40.4% over the tubewell water of the unamended field. Colour of the water samples of the amended fields was also darker than that of the unamended fields. The study indicated that the organic and inorganic ions added through the effluent could pose a serious threat to the groundwater quality if applied without proper monitoring.

Keywords: groundwater pollution, inorganic ions, leaching, nitrate, organic ions

Abbreviation: BOD, Biochemical oxygen demand; COD, Chemical oxygen demand; CPU, Chloroplatinate colour unit; EC, Electrical conductivity; MCL, Maximum concentration limit; PAR, Potassium adsorption ratio; PMDE, Post-methanated distillery effluent; QUEFTS, Quantitative Evaluation of Fertility of Tropical Soils-model; SAR, Sodium adsorption ratio; TDS, Total dissolved solids; USEPA, United State Environment Protection Agency

1. Introduction

Molasses-based distilleries are considered to be one of the most polluting agro-based industries due to generation of large amount of anaerobically digested, foul-smelling, coloured wastewater. The wastewater, called post-methanated distillery effluent (PMDE), has high chemical oxygen demand (COD, 45,000–50,000 mg L^{-1}) and biochemical oxygen demand (BOD, 5000–8000 mg L^{-1}) and is unsafe for disposal in watercourses (Joshi *et al.*, 1996). However, PMDE also contains appreciable amounts of macro- and micronutrients such as K, N, Fe, Cu, Zn and B, which are essential for plant growth. Several researchers (Joshi *et al.*, 1996;

Pathak *et al.*, 1999) have shown that the use of PMDE in agriculture for soil amendment or as a supplement to irrigation water in a judicious way improved crop growth, and physical, chemical and biological properties of soil. Pathak *et al.* (1999) suggested dilution of PMDE so as to bring down its BOD to 1000 mg L^{-1} before application in standing crop of rice and wheat for higher yield and improved soil properties. Deverajan *et al.* (1994) showed that PMDE could be safely used as liquid manure if applied at a rate of 125–250 cubic m ha^{-1}. However, non-judicious use of PMDE adversely affected crop growth and increased soil salinity (Jagdale and Sawant, 1979; Joshi *et al.*, 2000). Chonker *et al.* (2000) observed that use of PMDE should be discontinued intermittently for one to two crop-seasons to avoid the deteriorating effects on soil and groundwater quality.

Farmers in the adjoining areas of molasses-based distilleries often use the effluent for irrigating crops without considering its impact on the groundwater. Hence, an apprehension exists that the long-term use of PMDE in agricultural fields may pose a serious threat to the groundwater quality. Monitoring of groundwater beneath the effluent irrigation site is, therefore, important for maintaining its quality because of risk of leaching of organic and inorganic ions from effluent amendment. So far, no quantitative evaluation of leaching of salts from distillery effluent-irrigated fields in tropical countries have been made to assess the risk of groundwater pollution arising due to such a practice. The objective of the present study was to assess the impact of long-term application of PMDE on groundwater quality and apparent N budget.

2. Materials and Methods

2.1. EXPERIMENTAL SITE AND SOIL

Two on-farm experiments were conducted at village Gajraula in the district of Jyotiba Phule Nagar, Uttar Pradesh, India where a molasses-based distillery with the capacity of 300 KL alcohol per day is operating for the last 30 years. In the first field (PMDE-amended), located near the distillery, effluent was applied for irrigation for last 10 years at the rate of about 225 m^3 ha^{-1} as pre-sown irrigation. In the second field (unamended-control), located 1 km away from the distillery, no effluent was applied and irrigation was provided only from groundwater. Both the sites fall under the Indo-Gangetic plains of northwest India at 29°4′N latitude, 77°46′E longitude and an altitude of 237 m above mean sea level. The climate of the region is semi-arid subtropical, with dry hot summers and cold winters. Average annual rainfall is about 800 mm, 75% of which is received during monsoon season in July to September. Mean maximum and minimum temperatures are 34.0 and 24.1°C during the kharif season (July to October) and 22.6 and 6.7 °C during the rabi season (November to April). Rice-wheat is the dominant cropping system of the area. The groundwater hydrology of this region is characterized by unconfined and semi confined aquifers at a depth of about 20–25 m below ground level. The site is represented by alluvium

TABLE I
Physicochemical properties of soil of the experimental fields

Parameters	Amended field	Unamended field
Soil texture	Sandy loam	Sandy loam
Sand (%)	67.1	68.2
Silt (%)	20.0	18.0
Clay (%)	12.9	13.8
Bulk density ($g\,cm^{-3}$)	1.44	1.45
pH	7.6	7.1
EC ($dS\,m^{-1}$)	0.34	0.22
Organic carbon (%)	0.58	0.55
Total N (%)	0.05	0.04
Olsen P ($kg\ ha^{-1}$)	16.8	16.4
Amm. Ac. Exch. K ($kg\,ha^{-1}$)	370	190
SO_4^{2-} ($kg\,ha^{-1}$)	23	15
Na^+ ($kg\,ha^{-1}$)	146	123
Ca^{2+} ($kg\,ha^{-1}$)	1480	1447
Mg^{2+} ($kg\,ha^{-1}$)	412	325
SAR	4.8	4.1
PAR	12.0	6.4
CEC ($cmol\,kg^{-1}$)	10.6	10.4
Hydraulic conductivity ($cm\,h^{-1}$)	1.6	1.7
Porosity (%)	48	47

deposited during the Quaternary period. The geochemistry of the groundwater was same at both the locations. The sites were plain lands with negligible slope and the soils were Typic Ustochrepts, sandy loam in texture with high infiltration rate, low in organic carbon content and alkaline pH. The physicochemical properties of the soil of experimental fields are given in Table I.

2.2. TREATMENTS AND CROP MANAGEMENT

The PMDE used in the experiment had very high BOD and COD with EC 8.5 $dS\ m^{-1}$, and organic carbon, total N, total P and total K contents of 160000, 2100, 38 and 5838 $mg\ L^{-1}$, respectively (Table II).

In both the fields rice crop was grown during the monsoon season (called as kharif season in India) followed by wheat crop during the winter season (called rabi season). Urea was applied at a rate of 120 $kg\ N\ ha^{-1}$ in three splits at 60, 30 and 30 $kg\ N\ ha^{-1}$ to both the crops. Phosphorus (26.2 $kg\ P\ ha^{-1}$) and potassium (50 $kg\ K\ ha^{-1}$) were incorporated in the soil at the time of transplanting of rice and

TABLE II
Chemical composition of the post-methanated distillery effluent (PMDE)

Composition	Values
pH	7.8
EC (dS m^{-1})	8.5
TDS (mg L^{-1})	5440
BOD (mg L^{-1})	4830
COD (mg L^{-1})	31000
Organic carbon (mg L^{-1})	160000
N (mg L^{-1})	2100
P (mg L^{-1})	38
K (mg L^{-1})	5838
Na (mg L^{-1})	535
Ca (mg L^{-1})	332
Mg (mg L^{-1})	1274
Sodium adsorption ratio (SAR)	28
Potassium adsorption ratio (PAR)	206
Chloride (mg L^{-1})	2116
Sulphate (mg L^{-1})	4270
Zinc (mg L^{-1})	0.5

sowing of wheat using single super phosphate (SSP) and muriate of potash (KCl), respectively. In rice, 15 irrigations and in wheat 5 irrigations of 5–6 cm each were provided. Weeds, pests and diseases were controlled as required.

2.3. GROUNDWATER MONITORING

Groundwater samples from PMDE-amended field were collected from a tubewell (21-m deep) already located in the field as well as from three piezometers (16-m deep) installed in the field during the experiment. For the unamended-control field, groundwater samples were collected only from a tubewell (21 m deep) located in the field. Samples were collected throughout the study for two consecutive years (2001–2002) as per the following schedule: (1) on the day of field preparation, (2) once a week up to first 30 days of application of PMDE and (3) once a month during the rest of the period of the study. Samples were collected in polyethylene bottles (150 mL capacity) and were stored at 4 °C until further analysis.

Samples were analyzed for their physicochemical properties (colour, pH, EC, Na^+, K^+, Ca^{2+}, Mg^{2+}, NO_3^-, SO_4^{2-} and Cl^-) according to the standard methods given by American Public Health Association, Washington, DC (APHA, 1998).

Colour intensity in water samples was measured at the wavelength of 475 nm as per the APHA standard method using a spectrophotometer (EC, model GS 5701). Absorbance values were converted to chloroplatinate colour units (CPU) using the following equation.

$$\text{CPU} = 500A_2/A_1$$

where A_1 is the absorbance of 500 CPU standard solution and A_2 is the absorbance of the sample. Contents of Na^+ and K^+ in the samples were determined using a flame photometer (ELICO, CL361), Ca^{2+} and Mg^{2+} by Versinate titration method and Cl^-, PO_4^{3-}, NO_3^- and SO_4^{2-} using ion chromatograph (Metrohm 761, Compact IC). Total dissolved solids (TDS) were calculated by multiplying electrical conductivity (EC) with a factor of 640 (Falkiner and Smith, 1997). Sodium adsorption ratio (SAR) was calculated using the following formula.

$$\text{SAR} = \text{Na}/[\text{sqrt}(\text{Ca} + \text{Mg})/2]$$

2.4. ANALYSIS OF SOIL SAMPLES

Soil samples (0–30 cm depth) were collected from both the fields with the help of core sampler from five places. All the samples were mixed well and divided into three sub-samples and analyzed for the physicochemical properties.

2.5. ESTIMATION OF NITROGEN BUDGET

Annual apparent N budget for the unamended-control and PMDE amended fields was estimated using the following formula (Regmi *et al.*, 2002):

$$\text{N Balance} = \sum(\text{fertilizer N, manure N, rain N, biologically fixed N, irrigation-water N, N in seedling and seeds})$$

Among the various inputs N contents in mineral fertilizer, PMDE and irrigation water were measured in the present study. The contribution of 3.4 kg N ha^{-1} yr^{-1} with rainfall was based on the data by Mishra (1980) and Brown *et al.* (1999). Input from biological N fixation was considered at the rate of 10 kg N ha^{-1} during rice and 5 kg ha^{-1} during wheat (Brown *et al.*, 1999). In the PMDE treatment the biological N fixation was considered to be higher as amendment of organics is known to stimulate N_2 fixation (Roper and Ladha, 1995). The quantities of N added to the soil with rice seedlings and wheat seeds were obtained by considering their N content of 41 and 20 g kg^{-1}, respectively. Plant uptake of N was estimated using the QUEFTS (Quantitative Evaluation of Fertility of Tropical Soils) model calibrated for rice (Witt *et al.*, 1999) and wheat (Pathak *et al.*, 2003). Total losses of fertilizer N (volatilization, denitrification, and leaching) was taken to be 600 g kg^{-1} for rice (Brown *et al.*, 1999; Smaling and Fresco, 1993) and 500 g kg^{-1} for wheat (Tandon, 1994). Losses of N from soil were estimated based on data reported by Smaling and Fresco (1993) and Regmi *et al.* (2002).

2.6. STATISTICAL ANALYSES

Statistical analyses of the data were performed with SPSS for Windows (version 10.0) using paired '*t*-test'. Unless indicated otherwise, differences were considered only when significant at $P \leq 0.05$.

3. Results and Discussion

3.1. LEACHING OF NITRATE

Concentrations of NO_3^- in the groundwater samples from the piezometers and tubewell of the PMDE-amended field increased after effluent irrigation in rice crop (Figure 1). Before effluent irrigation, NO_3^- contents in the groundwater were 25.6 and 35.5 mg L^{-1}, in tubewell and piezometer samples, respectively, which increased to 58.6 and 71.0 mg L^{-1}, respectively, after irrigation. Nitrate

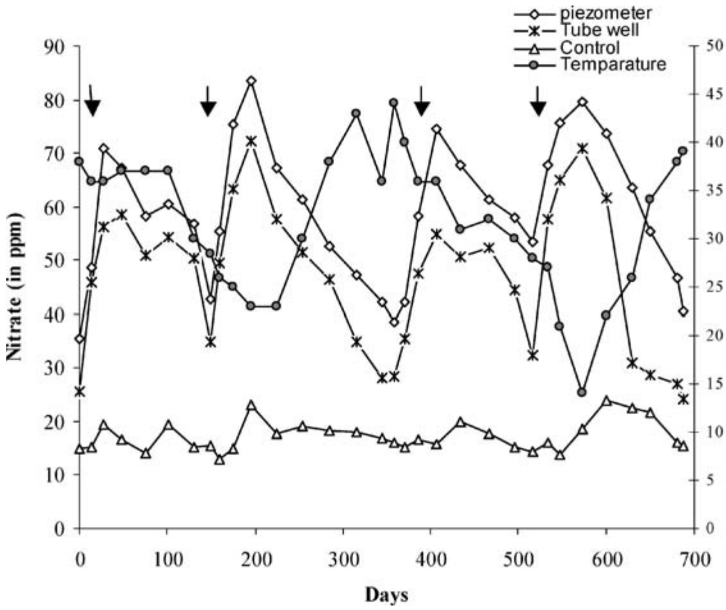

Figure 1. Nitrate concentration in groundwater samples collected from PMDE amended (piezometer and tubewell) and unamended fields (tubewell).

concentration in the groundwater samples from unamended field increased from 14.8 to 19.3 mg L^{-1} after irrigating with tubewell water. This increase was possibly due to high NO_3^- contents in the soil profile before rice crop, which was advectively transported downwards with percolating water. Higher NO_3^- content in the soil was responsible for higher NO_3^- loading of the groundwater below the amended field compared to that of the unamended field. Albus and Knighton (1998) found that irrigation caused a flush of NO_3–N to the shallow groundwater. Leaching of NO_3^- in unamended field was observed whenever N was applied through urea followed by irrigation.

Similar trends of NO_3^- leaching were observed during wheat crop in both the years. Nitrate content increased from 42.8 and 35.0 mg L^{-1} to 72.3 and 83.5 mg L^{-1} in tubewell and piezometer samples, respectively, of the amended plots, while NO_3^- content was 23.1 mg L^{-1} in the unamended field after irrigating with tubewell water. Larger amount of NO_3^- leaching were observed during the rabi season because of aerobic soil condition in which soil $NH_4{}^+$ is converted into NO_3^-, which is more susceptible to leach.

In the amended field NO_3^- contents of groundwater samples from piezometer were 35.5–83.5 and 38.5–79.7 mg L^{-1} in the year 2001–2002 and 2002–2003, respectively, whereas in the tubewell samples the values were 25.6–72.3 and 28.4–70.8 mg L^{-1}, respectively (Table III). A positive correlation was observed between NO_3^- contents in piezometer and tubewell samples ($r = 0.85$) of the amended field. However, NO_3^- contents were smaller in the tubewell samples compared to the piezometer samples (Table IV).

3.2. COLOUR

Colour in groundwater increased after initial application of effluent and irrigation (Figure 2). After the initial increase, colour decreased reaching a steady-state level. The colour units in groundwater samples from amended field were 75.76–174.24 and 25.76–71.52 CPU for piezometer and tubewell, respectively, in the first year, and 85.41–145.68 and 27.5–74.25 CPU for piezometer and tubewell, respectively, in the second year (Table III). Colour unit in groundwater samples from tubewell of unamended field was 3.78 (±0.01) CPU in both the years. Results indicated that the water samples collected from piezometer were more coloured in comparison to tubewell samples of amended field (Table IV).

3.3. PH AND EC

Groundwater samples from both the sites had alkaline pH ranging from 7.03 to 8.61. Average EC of the samples was 1.04 and 0.83 dS m^{-1} for piezometer and tubewell samples, respectively in the amended field. Total dissolved solids and anion/cation levels were higher in the field receiving PMDE than in the unamended field. It was

TABLE III

Groundwater quality of experimental and control field during 2001–2002

Treatment		NO_3 (mg L^{-1})	SO_4 (mg L^{-1})	K (mg L^{-1})	TDS (mg L^{-1})	Ca (mg L^{-1})	Mg (mg L^{-1})	Na (mg L^{-1})	Cl (mg L^{-1})	Colour	SAR
Year 2001											
Amended	Piezometer	57.8 (±3.2)[a]	74.8 (±6.1)	6.8 (±0.8)	708.0 (±45.9)	60.51 (±6.5)	12.0 (±19.1)	33.4 (±3.3)	43.0 (±4.8)	112.9 (±7.8)	3.6 (±0.4)
	Tubewell	48.8 (±3.2)	43.7 (±1.7)	4.0 (±0.6)	477.6 (±21.6)	41.1 (±3.6)	73.7 (±3.7)	22.1 (±1.7)	19.4 (±1.9)	54.3 (±3.4)	3.0 (±0.3)
Unamended	Tubewell	16.9 (±0.6)	17.3 (±0.3)	1.5 (±0.02)	314.4 (±3.6)	12.4 (±0.1)	25.4 (±0.2)	2.9 (±0.1)	13.0 (±0.1)	3.8 (±0.01)	0.7 (±0.02)
Year 2002											
Amended	Piezometer	59.8 (±3.3)	73.2 (±4.4)	6.0 (±0.9)	628.4 (±26.7)	67.3 (±5.3)	95.6 (±5.8)	40.3 (±2.7)	35.0 (±4.3)	107.3 (±5.5)	4.5 (±0.3)
	Tubewell	44.5 (±3.8)	49.1 (±2.3)	2.5 (±0.1)	590.8 (±30.9)	44.2 (±4.0)	69.7 (±3.8)	21.8 (±1.0)	23.0 (±1.7)	53.0 (±2.1)	3.0 (±0.2)
Unamended	Tubewell	17.4 (±0.8)	17.5 (±0.2)	1.5 (±0.02)	322.4 (±7.4)	13.1 (±0.3)	26.1 (±0.3)	2.8 (±0.1)	12.9 (±0.2)	3.8 (±0.01)	0.6 (±0.02)
Permissible limits[b]		50.0	250.0	6.0	500.0	200.0	–	175.0	250.0	colourless	–

[a] Values given in parenthesis is standard error of mean.

[b] Permissible limits for drinking water are as per United State Environment Protection Agency (USEPA).

TABLE IV

t-values between pair-wise water quality parameters

	Water quality parameters									
Treatments	NO_3	SO_4	Colour	TDS	K	Na	SAR	Ca	Mg	Cl
P1–T1[a]	11.16*	6.72*	9.44*	4.60*	4.33*	5.10*	1.90ns	3.56*	2.62*	6.58*
T1–C1	10.80*	17.63*	14.88*	7.62*	4.12*	11.30*	8.54*	8.01*	13.06*	3.28*
P2–T2	8.59*	6.79*	18.67*	1.13	4.08*	6.31*	5.02*	3.08*	3.01*	4.18*
T2–C2	7.13*	14.06*	11.99*	7.77*	8.52*	18.66*	15.02*	7.85*	11.61*	5.86*
P1–P2	1.20 ns	0.38 ns	1.26 ns	1.21 ns	1.15 ns	1.95*	2.46*	0.53 ns	1.44*	2.68*
T1–T2	2.29*	3.24*	0.52 ns	3.18*	2.74*	0.16 ns	0.22 ns	0.57 ns	0.64 ns	2.06 ns

[a] P1, T1, C1 are the samples from amended (piezometer), amended (tubewell) and unamended (tubewell) fields in the year 2001 and P2, T2, C2 are the samples in the year 2002, respectively.

* and ns are significant at 5% level and not significant, respectively, with paired *t*-test.

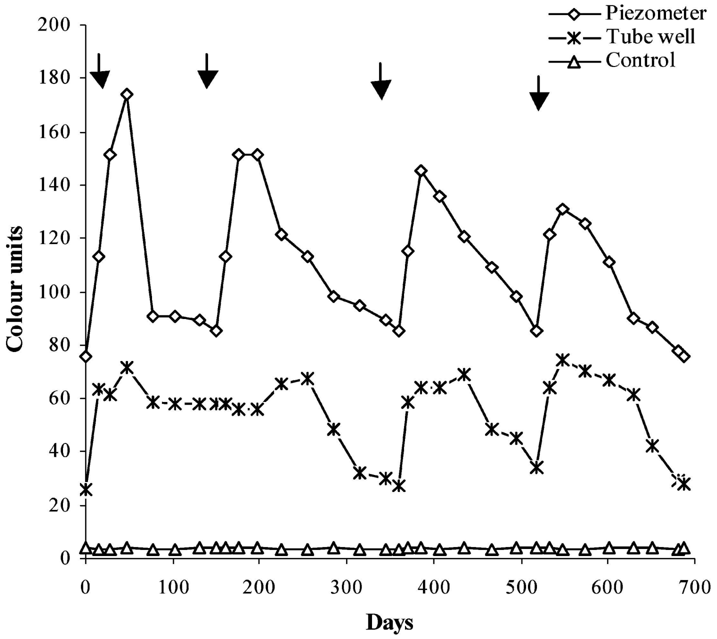

Figure 2. Colour units in groundwater samples collected from PMDE amended (piezometer and tubewell) and unamended fields (tubewell).

noted that TDS levels in the piezometer and tubewell samples in the amended field ranged from 441.6 to 998.4 mg L^{-1} and 332.8 to 812.8 mg L^{-1}, respectively. High salt content and TDS in groundwater due to effluent irrigation have also been reported by Joshi (1999).

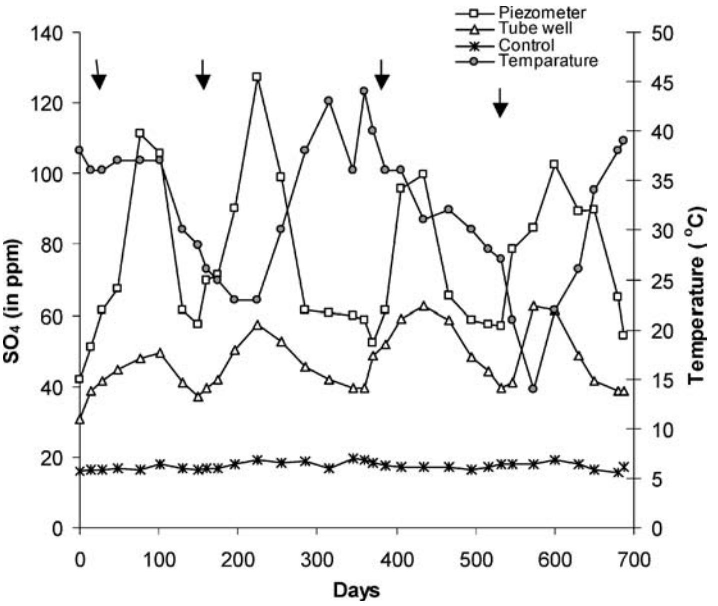

Figure 3. Sulphate concentration in groundwater samples collected from PMDE amended (piezometer and tubewell) and unamended fields (tubewell).

3.4. LEACHING OF OTHER NUTRIENTS

Increased concentrations of SO_4^{2-} in groundwater due to PMDE amendment was observed in the study (Table III) but it was within the permissible limits established by USEPA (MCL 250 ppm SO_4^{2-}). The trend of SO_4^{2-} leaching was similar to NO_3^- leaching (Figure 3).

Elevated concentrations of chloride (Figure 4) and potassium (Figure 5) in groundwater samples were also observed in amended field compared to that of the unamended field (Table III). Sodium concentration of the groundwater samples from amended piezometer increased over time from initial 36.2 to 68.8 mg L^{-1}. Sodium adsorption ratio in the tubewell samples increased from 0.6 in the unamended field to 4.8 in the amended field. Application of paper and pulp mill effluent in New Zealand increased SAR from 2 to 16 and also increased Na concentration in groundwater (Johnson and Ryder, 1988).

3.5. APPARENT N BALANCE

The apparent N balance was positive ranging from 4 kg N ha^{-1} yr^{-1} in the unamended field to 380 kg N ha^{-1} yr^{-1} in PMDE amended field. In case of PMDE amendment the balance was positive in both rice and wheat crops whereas in unamended field it was positive in rice and negative in wheat crop. Such positive

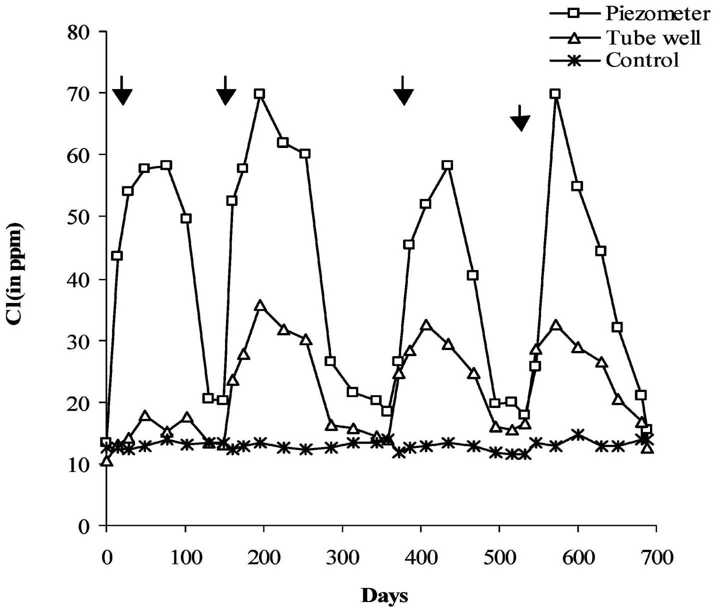

Figure 4. Chloride concentration in groundwater samples collected from PMDE amended (piezometer and tubewell) and unamended fields (tubewell).

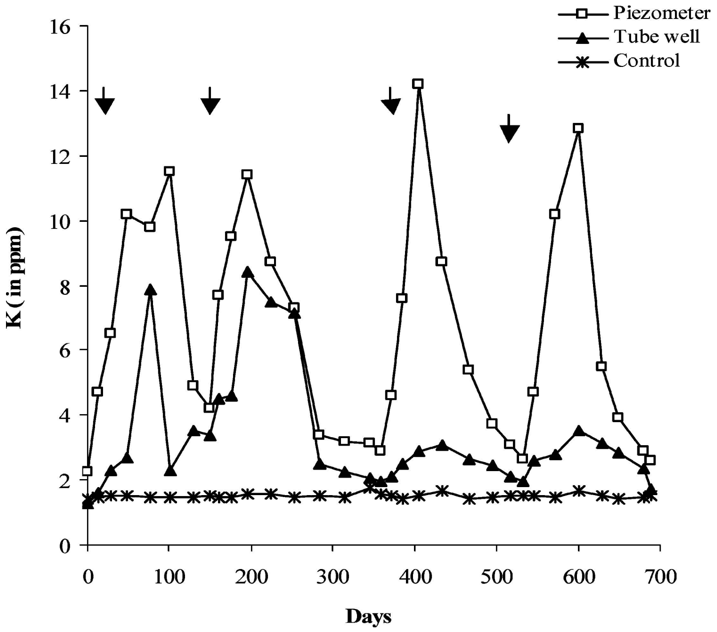

Figure 5. Potassium concentration in groundwater samples collected from PMDE amended (piezometer and tubewell) and unamended fields (tubewell). Downward arrows indicate effluent application.

TABLE V

Correlation coefficient between quality parameter pairs of the water samples from piezometer and tubewell

Quality parameter pairs	Coefficient of correlation (r)	
	Piezometer	Tubewell
$Na^+–NO_3^-$	0.01	0.26
$Na^+–SO_4^{--}$	−0.04	0.24
$Na^+–Cl^-$	−0.06	0.11
$Na^+–TDS$	0.055	0.18
$K^+–NO_3^-$	0.72^{**}	0.46^{**}
$K^+–SO_4^{--}$	0.71^{**}	0.33
$K^+–Cl^-$	0.86^{**}	0.50^{**}
$K^+–TDS$	−0.03	0.013
$Mg^{++}–SO_4^{--}$	0.45^{**}	0.12
$Mg^{++}–Cl^-$	0.37^{*}	0.22
$Ca^{++}–Cl^-$	−0.28	0.31
$TDS–NO_3^-$	0.19	0.48^{*}
$TDS–SO_4^{--}$	0.11	0.48^{**}
$TDS–Cl^-$	0.21	0.69^{**}

*,** are significant at 5% and 1% probability levels, respectively.

balances of N in the amended field were responsible for NO_3^- leaching into the groundwater whenever there is irrigation or rains.

3.6. CORRELATION BETWEEN QUALITY PARAMETER PAIRS OF WATER SAMPLES FROM PIEZOMETER AND TUBEWELL

Correlation between quality parameter pairs of groundwater samples from piezometer and tubewell in amended field are shown in Table V. Significant correlation existed between K^+/NO_3^-; K^+/Cl^-; K^+/SO_4^{2-}; Mg^{2+}/Cl^-; Mg^{2+}/SO_4^{2-} in piezometer samples and between K^+/NO_3^-; K^+/Cl^-; TDS/SO_4^{2-}; TDS/Cl^-; TDS/NO_3^- in tubewell samples suggesting that there is a common source of pollution at both the depths in amended field. There was low correlation between other quality parameter pairs of both the sources.

The study indicated that long-term indiscriminate use of PMDE could lead to significant leaching of inorganic salts. Although leaching of salts has the potential to affect the quality of groundwater, the actual impact will depend on the rate of recharge of groundwater, initial status of groundwater quality and agro-management practices followed in the area. Therefore, PMDE must be applied judiciously according to crop requirements and soil fertility status to prevent contamination of shallow groundwater with nitrate and other toxic ions.

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