

Changes in the recoverable fractions during successive cycles of aquifer storage and recovery system in North-West India

Y. S. Saharawat^{1*}, R. S. Malik², B. S. Jhorar², T. Streck³, Neelam Chaudhary⁴ and M. L. Jat⁴

¹International Rice Research Institute, 1st Floor, NASC Complex, DPS Marg, Pusa, New Delhi 110 012, India

²Department of Soil Science, CCS HAU, Hisar 125 004, India

³Biogeophysics Section, Universitat Hohenheim, Stuttgart, Germany

⁴CIMMYT India, Ground Floor, NASC Complex, DPS Marg, Pusa, New Delhi 110 012, India

Aquifer storage and recovery (ASR) is an effective method of recharging reclaimed or surplus surface water into confined or semi-confined permeable formations for later extraction. A key factor in the long-term viability of ASR is the extent of mineral interaction between two dissimilar water types, their recoverable fractions and consequent impact on water quality and aquifer stability. An ASR well study was undertaken in the semi-arid region of northern India to assess the technical viability, environmental sustainability and commercial/economic feasibility of the ASR system in a generic sense. 300 m³ of canal water was injected in each recharge cycle of brackish water and 100% of the mixture was extracted during each recovery cycle of the experiment. The study revealed that groundwater quality of the recovered water was better than that of the native water and recovery percentage of the recharged water (EC > 2 dS m⁻¹) increased from 27% to 42% in the successive recovery cycles. The potassium concentration in the recovered water was greater than that of the injected water as a result of potassium release from clay minerals in the aquifer. Borate was also released from the aquifer due to tourmaline dissolution and desorption from clay minerals under lower pH conditions caused by surface water injection.

Keywords: Aquifer storage and recovery, groundwater quality, hydro-geochemistry, semi-arid regions.

AQUIFER storage and recovery (ASR) is a relatively new water resource management technology, which has been put to a wide range of uses¹, including the improvement of groundwater quality for irrigation²⁻⁴, particularly in arid, semiarid and coastal areas. ASR involves the subsurface freshwater recharge and subsequent extraction during periods of water deficit or high demand through successive cycles to meet crops/plantation water requirements. This has not been a part of traditional practices in water

resource management because success relies on an emerging understanding of the subsurface process, especially in the semi-arid areas. In semi-arid regions, the surplus rain, canal and river water available during wet period may be recharged to improve the quality of native brackish aquifers for subsequent irrigation to increase crop productivity in dry periods.

More than 53% of the groundwater in Haryana (India) is brackish (EC > 2 dS m⁻¹) (ref. 5) and in other parts of the country, this number ranges from 32 to 83% (ref. 6). Out of the total surface water potential of Haryana (14.8 × 10⁹ m³/annum) about 36% goes unutilized⁷. This suggests that other semi-arid regions also could have a similar large potential for utilizing the excess fresh surface water to improve the quality of underground brackish water using the ASR technology.

Most wells in northern India are of cavity type and did not clog when recharged with large quantities of fresh water (900 mg l⁻¹; ref. 5). Clogging has been reported to be the major problem in most of the filter type ASR wells⁷⁻¹⁰. Cavity wells are shallow wells installed in aquifers (15 to 100 m deep) where an empty space or cavity is formed below the impermeable layer¹¹.

The geochemistry of ASR systems is complex and is still being studied. Prior knowledge of geochemical reactions occurring in the aquifers during mixing of recharge water with groundwater of different mineral composition and pH, and possible reactions with the mineral assemblage of the host rock would help in installation, operation and sustenance of an ASR system. An understanding of the precipitation of iron, manganese and arsenic^{1,12-14} in the aquifer by injecting pH optimized water may be utilized to reduce the impact of these elements on drinking water and the clogging of ASR wells. Similarly, the knowledge of enhanced dissolution of calcite minerals in the aquifers can be used to increase the hydraulic conductivity of the aquifer^{9,15,16}.

The present study was therefore, initiated at the Regional Research Station Balsamand of Chaudhary Charan Singh Haryana Agricultural University (CCS HAU) Hisar, Haryana with objectives to quantify (1) the mixing and physicochemical interactions between native and recharged water and (2) the effect of freshwater recharge on quality improvement and nutritional value of recovered water from cavity type brackish ASR well for irrigation purpose.

The recovery percentage I is defined as the percentage recovered water volume V_r at any recovery time t_r to the recharged volume V_i

$$I = \frac{\int_{t_{i1}}^{t_{r2}} q_r(t) dt}{\int_{t_{i1}}^{t_{i2}} q_i(t) dt} \times 100 = \left[\frac{V_r}{V_i} \right] \times 100, \quad (1)$$

*For correspondence. (e-mail: ysaharawat@cgiar.org)

where t_{i1} = time when recharge starts, t_{i2} = time when recharge ends, t_{r1} = time that recovery starts, t_{r2} = time when recovery ends, $q_r(t)$ = recovery rate as a function of time, $q_i(t)$ = recharge rate as a function of time, V_r = volume recovered between recovery time t_{r1} and t_{r2} and V_i = volume recharged between recharging time t_{i1} and t_{i2} .

The percentage of native water in the cumulative volume of recovered water, for any of the quality parameters, was defined as consistent with the definition used by Pavelic *et al.*¹⁶ and Ragone and Vecchioli¹⁷

$$M(t) = \frac{C_{rw}(t) - C_i}{C_n - C_i} \times 100, \quad (2)$$

where $C_{rw}(t)$ is the average concentration of a given parameter in the cumulative recovered volume of water V_r ; C_i and C_n are concentrations of the same parameter in recharged and native water. Concentration $C_{rw}(t)$ in cumulative water volume is indicative of the quality change in recovered water stored in the tank before use and can be estimated as:

$$C_{rw}(t) = \frac{\int_{t_{r1}}^{t_{r2}} C_r(t) q_r(t) dt}{\int_{t_{i1}}^{t_{i2}} q_i(t) dt} = \frac{\sum C_r(t) V_r}{\sum V_r}, \quad (3)$$

where C_r is the instantaneous concentration of a given parameter as a function of time t in the instantaneous recovered water sample V_r .

Chloride is a conservative ion which is not supposed to undergo any precipitation, dissolution, adsorption and ion exchange in the soil water system; therefore, it is used as an indicator ion for quantifying the simple mixing process between native and recharged water. The native water percentage C_x in the cumulative recovered water as explained in the previous section can also be used to quantify the physical and chemical processes. Let C_x for chloride at 100% recovery be C_{cl} . If a water quality parameter that shows a C_x value close to C_{cl} value (critical limit assumed is within 10% of C_{cl} value)^{1,18} then the parameter is considered to have gone through the process of mixing only (no physical and chemical reaction). However, C_x value beyond the range $C_{cl} \pm 0.1 \times C_{cl}$ means that some other interactions have taken place in addition to simple mixing. Depending on whether C_{cl} is more than 1.10 C_{cl} or less than 0.9 C_{cl} and the concentration of the particular parameter in the native groundwater $C_n(X)$ and the recharged water $C_i(X)$, different physical and chemical processes will occur and lead to production or consumption of an ion.

The total amount of salt/parameter (TA) present in the recovered volume of water (V_r) can be estimated as:

$$TA(t) = \int_{t_{r1}}^{t_{r2}} C_r(t) q_r(t) dt = V_r \times C_{rw}(t). \quad (4)$$

The amount of salt/parameter due to mixing (MA) is estimated as:

$$MA(t) = C_n \left[V_r \frac{C_{cl}}{100} \right] + C_i \left[V_r - V_r \frac{C_{cl}}{100} \right]. \quad (5)$$

The amount of salt/parameter produced/consumed (IA) due to geophysical and chemical interaction is given by the difference of TA and MA.

Bicarbonate (HCO_3^-) exists in equilibrium with carbonic acid (H_2CO_3), which in turn can be converted to carbon dioxide and water.



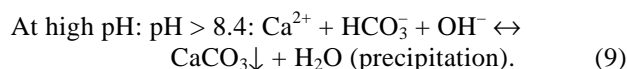
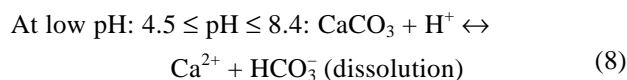
This means that the direction of a reaction would depend upon pH difference between recharged and native waters. The relative amount of carbonic acid and bicarbonate ion will be determined by the pH of the equilibrium solution.

Similarly, borate ($H_2BO_3^-$) exists in equilibrium with boric acid (H_3BO_3).



The direction of the reaction depends upon the pH difference between recharged and native waters.

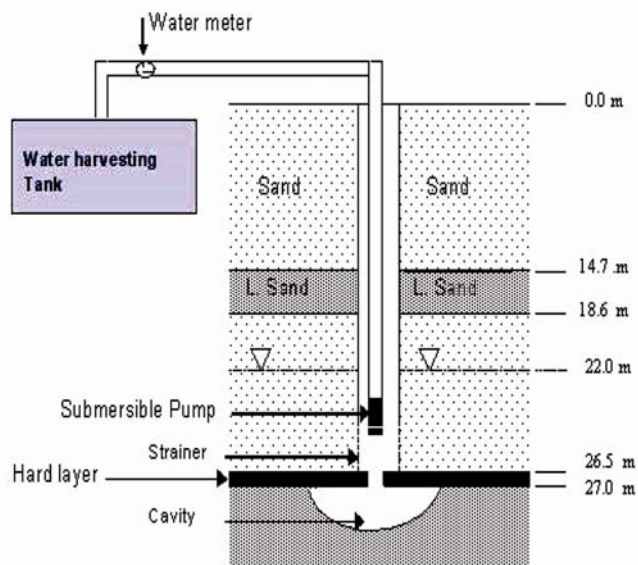
Dissolution and precipitation of $CaCO_3$ are common phenomena. The solubility of $CaCO_3$ is affected by different factors such as pH, CO_2 , temperature, organic activity, etc. Effect of pH on the solubility of $CaCO_3$ may be described by the Piper diagram¹⁹.



Unconsolidated material deposits in Haryana are very deep (>200 m) and consist of both eolian and alluvial sediments deposited by rivers emanating from the Himalayas. These materials range in composition from coarse to silty, clayey fine sand and are known to contain illite clay²⁰. The underlying bedrock consists of sandstone and limestone down to a depth of roughly 450 m. Groundwater within the unconsolidated deposits occurs largely under water table conditions²¹, the depth to the groundwater flow from the bedrock into the shallow alluvial aquifers. Well yields from shallow aquifers range between 1.5 and 30 l s⁻¹, and salinity of the water pumped from these wells varies from less than 2 dS m⁻¹ to more

Table 1. Relevant soil physicochemical properties of Hisar ASR site

Depth (m)	EC (dS m ⁻¹)	pH	Texture	CaCO ₃ (%)	Calcite concretion (%)	Gypsum (%)
0.0–14.7	3.1	8.8	Sand	1.4	12.0	0.014
14.7–18.6	2.9	8.3	Loamy sand	1.5	33	0.015
18.6–26.5	2.2	8.2	Sand	7.6	17.6	0.020
26.5–27.0	2.7	8.8	Sandy loam	0.7	3.5	0.015
27.0–30.0	2.6	8.9	Loamy sand	1.4	3.8	0.014

**Figure 1.** Schematic diagram of the ASR well.

than 30 dS m⁻¹. Deeper wells draw from the sandstone and limestone aquifers, which yield up to 150 l s⁻¹.

An ASR site of highly brackish native water was selected at Regional Research Station, Balsamand of CCS Hararyana Agricultural University, 26 km away from Hisar, where an irrigation cavity type well was installed within the shallow, alluvial aquifer. The relevant site characteristics are given in Table 1. The diameter of inner and outer pipes of the ASR well was 0.025 m and 0.075 m respectively (Figure 1). In March 2001, good quality (EC = 0.35 dS/m) tubewell water (300 m³) was recharged by gravity into cavity type ASR well, employing a siphon system during each cycle to study the effect of successive number of cycles. Recovery of the water started immediately after recharge without any storage time.

Soil samples from different layers taken during the installation of piezometers, were oven dried and ground gently with pestle–mortar. The fraction remaining (concretions) on a 2 mm sieve was analysed for calcite. The sediments passed through the sieve was analysed for different physicochemical properties. The relevant physicochemical properties up to the aquifer are given in Table 1. Samples of recovery water as a function of recovery time and of recharged and native water were analysed for

temperature, organic carbon (OC)²², cations Na⁺, K⁺ (flame photometer), Ca²⁺, Mg²⁺ (versenate method, ethylene diamine tetraacetic acid (EDTA)], NH₄⁺, Zn²⁺ and anions CO₃²⁻, HCO₃⁻ (acidimetric method), Cl⁻ (potassium chromate method), SO₄²⁻ and BO₃⁻ (calometric method). Per cent error in ionic mass balance E_m was calculated¹ as

$$E_m (\%) = 100 \left[\frac{\sum (EC_c - EC_a)}{\sum (EC_c + EC_a)/2} \right], \quad (10)$$

where EC_c and EC_a are cation and anion concentrations in mmol_c L⁻¹.

Recharge and recovery rates were fairly constant (3.12 m³ h⁻¹) during all the cycles. Clogging was not observed at the site, as recovery rates remained unaffected with successive ASR cycles.

Cationic and anionic composition, EC and pH of recharged water, native groundwater and recovered water along with average concentration (C_{rw}) at 50% recovery and corresponding cumulative mixing percentage M are presented in Table 2. Less than 10% error in charge balance E_m (eq. 10, Table 2) validate the laboratory analyses for meaningful interpretation of aquifer geophysical interactions.

As chloride does not participate in geophysical interactions¹, it was taken as an indicator ion for quantifying the mixing process between native and recharged water. The chloride percentage C_{cl} in the cumulative recovered water volume at any recovery percentage I quantifies simple mixing process as the fraction of native water mixed in recovered water. Simple mixing as represented by chloride in native water percentage in recovered water at 50% recovery chloride C_{cl} decreased linearly with successive cycles as

$$C_{cl} = -2.82x + 11.15; \quad r^2 = 0.82. \quad (11)$$

This is because recharged water left in aquifer after each cycle acts as a buffer zone that restricts the direct mixing of recharged water with the native water, and this leads to successive decrease in the proportion of native water in the recovered water with successive cycles.

A comparison of each parameter of chloride in the native water percentages in the recovered water was at 50% recovery and showed that bicarbonate, borate,

Table 2. Concentration (mmol l⁻¹) of different quality parameters in native (C_n) and injected (C_i) water and cumulative recovered water C_{rw}* at 50% recovery

Parameter	C _n	C _i	C _{rw} (C _x)		
			I	II	III
EC (dS m ⁻¹)	28.7	0.2545	3.57 (13.23)	4.83 (16.56)	4.05 (15.18)
Cl (mmol l ⁻¹)	277	1.50	26.56 (9.09)	12.56 (4.01)	11.02 (3.45)
SO ₄ ²⁻ (mmol l ⁻¹)	0.63	0.008	0.15 (10.91)	0.069 (4.21)	0.046 (2.54)
HCO ₃ ⁻ (mmol l ⁻¹)	5.00	1.75	2.70 (35.62)	2.60 (26.35)	2.51 (23.52)
BO ₃ ⁻ (mmol l ⁻¹)	0.045	0.015	0.092 (22.85)	0.054 (11.60)	0.052 (11.18)
NO ₃ ⁻ (mmol l ⁻¹)	2.00	0.04	0.83 (40.43)	0.55 (26.36)	0.49 (23.17)
Na ⁺ (mmol l ⁻¹)	150	0.35	15.13 (9.8)	7.05 (4.48)	6.14 (3.86)
K ⁺ (mmol l ⁻¹)	1.30	0.10	0.30 (17.0)	0.23 (10.97)	0.20 (8.61)
Ca ²⁺ (mmol l ⁻¹)	17.4	0.63	3.57 (6.91)	2.6 (4.2)	2.4 (3.57)
Mg ²⁺ (mmol l ⁻¹)	62.9	0.98	16.90 (10.07)	7.16 (4.21)	6.23 (3.45)
Em (%)	4.87	8.34	-4.42	-9.56	-8.90
pH	8.90	7.40	7.50	7.62	7.74

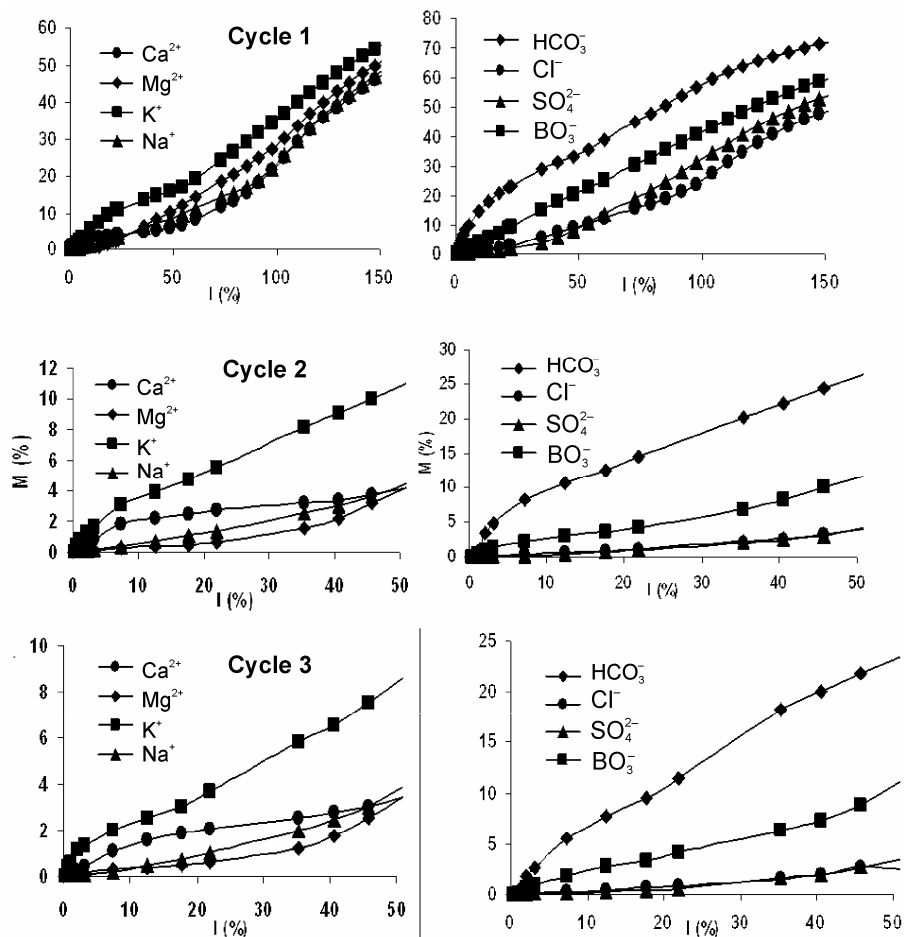


Figure 2. Amount of quality parameters recovered with time in the recovered water in all ASR cycles.

nitrate and potassium of the recharged water have been affected most by geochemical reactions between the native groundwater and recharged water (Table 2). Other parameters in the recovery water were mainly affected by simple mixing between native groundwater and recharged water.

The mixing showed an increase in mixing (*M*) recovery (*I*) for all quality parameters for all ASR cycles in Figure 2. This means that the water recovered was a mixture of recharged water and native groundwater and the proportion of native groundwater increased with recovery, percentage showing increasing mixing *M* as the recovered

water is withdrawn radially away from the ASR well. Mixing curves of M versus I showed that M increased linearly. Dispersion and regional movement of the recharged water bubble may have increased the mixing. The dependence of C_x on these factors was also emphasized by Pavelic *et al.*¹⁶.

The natural groundwater chemistry is dominated by sodium and chloride. Native groundwater salinity is approximately 14245 mg l^{-1} ($\text{EC} = 24.7 \text{ dS/m}$) and is therefore unsuitable for irrigation. Equilibrium with the limestone aquifer leads to significant concentrations of calcium and bicarbonate.

At 100% recovery percentage Ca^{2+} and HCO_3^- in the cumulative recovered water volume were much higher than that of simple mixing C_{cl} value with all ASR cycles. This means that if only simple mixing had occurred, the concentration of HCO_3^- in recovered water would have been within $\pm 10\%$ of C_{cl} . Therefore, it was a case, where $M(\text{HCO}_3^-) > 1.10 C_{cl}$ and $C_n(\text{HCO}_3^-) > C_i(\text{HCO}_3^-)$, suggesting that calcite (CaCO_3) is dissolving. The relatively low pH value of recharged water ($\text{pH} = 7.40$) as compared to that of native groundwater water ($\text{pH} = 8.95$) may have caused dissolution of calcite (CaCO_3) present in the aquifer material (Table 2) to form Ca^{2+} and HCO_3^- following eq. (6) as



This means that one mole of calcite mineral (equal to 100 g) would produce one mole of Ca^{2+} and two moles of HCO_3^- . Comparing HCO_3^- production with Ca^{2+} production from 150 m^3 of recovered water during the first ASR cycle, it was found that HCO_3^- and Ca^{2+} were produced almost in a ratio of 2 : 1 in the successive cycles. The dissolution and interaction amount IA HCO_3^- at 50% recovery decreased with successive cycles because of the decreasing cumulative native water percentage in the recovered water with increasing buffer storage in native water of recharged water. Nevertheless their production proportion to their cumulative mixing also followed the decreasing pattern with successive cycles.

K^+ and BO_3^- concentrations in the recovered water were much higher than the C_{cl} (Table 2). This shows that potassium and borate were released from the aquifer clay minerals, possibly due to freshening of the brackish groundwater. It is likely that potassium was released from its adsorbed/non-exchangeable state as a result of increased hydraulic pressure created by the recharge process. The relatively low pH of the recharge water (7.40) as compared to that of native groundwater pH (8.95) may have caused the desorption process in the aquifer and dissolution of borate from tourmaline, a boron-bearing mineral. During the first ASR cycle, 283.3 mol_c (14 kg) of potassium was released in recovery water. Potassium release decreased with increasing successive cycles, as in the case of calcite dissolution. Potassium

release decreased from 283.3 mol_c to 33.82 mol_c in three successive ASR cycles. Similarly the interaction amount for borate decreased with successive ASR cycles (Table 2). Malik *et al.*⁵ also reported K release in ASR of semi-arid region.

Groundwater quality of the recovered water in terms of electrical conductivity EC was better than that of native water. The proportion of native groundwater in recovered water increased with recovery percentage I . It implied that the first water has much better quality than water at the end of the season. This would be beneficial for the crops as the crops are more sensitive at the earlier stages of growth.

Recovery efficiency (RE) is defined as the recovery percentage I at target time to meet the target cumulative EC_{rw} of the recovered water (2 dS m^{-1}). RE increased linearly from 27 to 42% with successive ASR cycles.

$$\text{RE} = 6.1\text{SC} + 34.3, \quad r^2 = 0.99. \quad (14)$$

Increased RE with successive ASR cycles was due to decreased mixing with increasing buffer storage volume.

The present study showed that all the quality parameters (cations and anion) increased with time in the recovered water in all ASR cycles. There was a decrease in the simple mixing at 50% recovery C_{cl} with successive ASR cycles. In all successive ASR cycles, the dissolution of calcite was in a ratio of 2 : 1 of Ca^{2+} and HCO_3^- . Release of Ca^{2+} and HCO_3^- was from dissolution of calcite and K^+ from clay minerals and borate due to desorption process at higher pH in the aquifer with successive ASR cycles. With increasing number of ASR cycles, recovery efficiency at target EC_{rw} of 2 dS m^{-1} increased linearly from 27 to 42%, showing a buffer volume of good quality water in aquifers.

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