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DIFFUSE AGRICULTURAL NITRATE POLLUTION OF GROUNDWATERS IN INDIA

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ABSTRACT

Nitrate pollution of groundwater due to urban waste and industrial effluents usually centres around cities. This study has shown that nitrate levels in groundwater over vast agricultural areas can be correlated with intensive irrigated agriculture, corresponding use of nitrogenous fertilizers and groundwater development, and consequent diffuse agricultural pollution has already endangered the safety of potable groundwater for future generations in both rural and urban areas. Chemical and bacterial treatment of groundwater for nitrate removal relies on advanced technology and is considered costly even in the developed world. In a country like India where economic resources are inadequate, action on the suggested preventive measures may be taken without delay at this stage when alarming trends have been recorded. © 1999 IAWQ Published by Elsevier Science Ltd. All rights reserved

KEYWORDS

Diffuse pollution; agricultural pollution; nitrate pollution; groundwater; fertilizer consumption.

INTRODUCTION

Groundwater, due to its relative purity, enjoys a privileged place as a potable water source in India. The merits of groundwater over surface water are: little or no need of treatment to ensure potable quality, easy availability, and natural annual replenishment by rainfall. Unfortunately, the discharge of agricultural, industrial and domestic waste for the past several decades has been causing leaching of hazardous organic and inorganic chemicals into groundwater aquifers, thereby posing destruction of the aforesaid virtues. Nitrate is one such "anthropogenic pollutant" contributed by nitrogenous fertilizers, industrial effluents, and human and animal wastes through the biochemical activity of nitrifying bacteria, such as *Nitrosomonas* and *Nitrobacter.* In pollution-free areas it is produced by soil organic matter decaying through these bacteria, the intensity of activity depending on soil type, vegetation and climate. Recent studies in Australian arid zones have shown that bacteria associated with certain soil termites can cause considerable nitrate pollution of shallow groundwater under flash desert precipitation events (Barnes *et al.*, 1992). No such information has yet been gathered from Indian arid zones. Denitrifying bacteria can convert the nitrate back to N_2O and nitrogen by anaerobic reduction. But in the absence of such a process, nitrate infiltrating deep into aquifers may remain as such for a long time.

NITRIFYING - DENITRIFYING REACTIONS

Urea (NH_2CONH_2) is the most common nitrogenous fertilizer applied to agricultural fields in India and is also a major intermediate product of protein metabolism. Urobacteria and other microbes hydrolyse urea by the following reaction.

$$CO(NH_2)_2+2H_2O = (NH_4)_2CO_3$$

This reaction can take place in both aerobic and anaerobic conditions. Ammonium carbonate, being a salt of a weak acid and a weak base, easily hydrolyses:

$$(NH_4)_2CO_3 + HOH = NH_4(OH) + NH_4HCO_3$$

Dissociation of ammonium hydroxide is expressed by the equilibrium:

$$NH_4OH = NH_3 + H_2O$$

The ammonia so derived is bacterially oxidized in two distinct phases under the action of microorganisms *Nitrosomonas* and *Nitrobacter*. The first step is activated by *Nitrosomonas:*

$$2NH_3 + 3O_2 \Rightarrow 2HNO_2+2H_2O + 158 \text{ kilocalories}$$

The second step takes place in the presence of *Nitrobacter*:

$$2HNO_2 + O_2 \Rightarrow 2HNO_3 + 43 \text{ kilocalories}$$

Both the above reactions are exothermic, very slow and are mildly temperature dependent. Conversion of 10 mg of ammonium nitrogen into nitrites may take 15 days and 10 mg of nitrites may get nitrated in 40 days. The variation of temperature in the range 9 to 26°C does not result in a change in the speed of reaction but it is slowed down as the temperature drops below 9°C. At 6°C the reaction is substantially impeded and at 0°C nitrification practically stops (Voznaya, 1981). In the tropical to sub-tropical Indian climate, nitrification proceeds at relatively faster rates compared to those in the temperate climate of western countries.

The reverse process, i.e. reduction of nitrate to nitrite and finally to free nitrogen gas, is induced by facultative anaerobes and denitrifying bacteria. Denitrification occurs in the presence of nitrogen free organic compounds like carbohydrates, cellulose, salts of volatile fatty acids, etc., which are oxidised by the oxygen liberated from nitrates providing energy for the reaction. This process which operates in the absence of free oxygen and in the presence of organic matter can be expressed by the following generalized reaction:

$$4KNO_3 + 5C_{org} \Rightarrow 2K_2CO_3 + 3CO3 + 2N_2$$

The process of denitrification can also take place in the absence of organic matter when sulphur is present:

$$6KNO_3 + 5S + 2CaCO_3 = 3K2SO_4 + 2CaSO_4 + 2CO_2 + 3N_2$$

(*Thiobacillus denitrificans*) (Sulphate deposits in sewage structures)

The above reaction is induced by *Thiobacterium denitrificans.*

NITRATE LEVELS IN TERRESTRIAL WATERS

Terrestrial waters include polar ice-caps, water frozen at high altitude, water stored in lakes and rivers and subsurface rock formations (groundwater). Nitrate levels in relatively pollution-free areas of continents such as high altitude lakes and rivers and snow-clad mountains helps in understanding the anthropogenic nature of this pollutant in groundwaters (Table 1). In central Himalayan snow and ice, the nitrate level is about 0.5

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mg/l. The average nitrate level in world rivers is about 1 mg/l which is close to its content in Himalayan rivers, e.g. the Bhagirathi and the Alaknanda (Ganga source waters) and in the Ganga at Rishikesh before it enters the plains. The slight increase in NO_3 in river waters compared to that of frozen water may be due to contributions from biochemically derived soil nitrate. In Lake Dal of Kashmir, nitrate was below detectable range (i.e. < 0.01 mg/l) in 11 out of 19 samples (Handa *et al.*, 1991). However concentrations up to 6.8 mg/l were noticed in one of the samples. No nitrate was detected in rainwater around Lake Dal. It thus emerges that in a relatively pure system, surface water contains less than 1.0 mg/l NO_3, and its higher concentrations in groundwater may therefore reflect anthropogenic or geological contributions. In oceans, which are the ultimate sink for terrestrial waters, the average NO_3 level is 0.67 mg/l (Mason and Moore, 1985). This is slightly lower than in average river water indicating possible biochemical removal by marine plankton. In the USA nitrate in terrestrial water ranges from 1 to 20 mg/l, the higher content being in groundwaters.

Under natural conditions anaerobic denitrification of NO_3 into nitrous oxide (N_2O) balances the downward percolation of excess NO_3 through soil. The groundwater nitrate level at which that balance occurs varies with geology and local environment but seldom exceeds 12-13 mg/l as NO_3 (about 3 mg/l nitrate nitrogen) and is often only one tenth of that amount. The United States EPA therefore chooses 3 mg/l NO_3-N (13.3 mg/l as NO_3) as an approximate level beyond which human activities may be taken to be contributing nitrogenous compounds to the water (Anonymous, 1987).

Table 1. Nitrate in Indian terrestrial waters in relatively pollution free areas

Locality	Nitrate (mg/l)
Central Himalayan snow*	0.496
Central Himalayan ice*	0.436
Himalayan rivers*	
Bhagirathi	0.310 to 0.992
Alaknanda	0.992
Ganga (at Rishikesh)	0.806
Lake Dal water**	1.07
World average river water***	1.00
World average ocean water***	0.67

* Calculated by author after Sarin *et al.* (1992).
** Average of 19 samples reported by Handa *et al.* (1991).
*** After Mason and Moore (1985).

ADVERSE EFFECTS OF NITRATE AND NITRATE STANDARDS FOR POTABLE WATER

Nitrate concentration in drinking water at 10-45 mg/l or more is considered to be carcinogenic and a causative factor for "blue" babies. The functioning of the central nervous system (CNS) and cardiovascular system (CVS) may also be adversely affected by nitrate-rich water. The World Health Organization (WHO) has recommended the limit of 10 mg/l nitrate-nitrogen (NO_3-N) for drinking water which is equivalent to about 45 mg/l of nitrate (NO_3) and the same is also accepted in India by the ICMR.

NITRATE POLLUTION OF GROUNDWATER IN INDIA: THE STATE OF AWARENESS AND INFORMATION

Some cases of high NO_3 in groundwaters in India has been reported by Handa (1975), Kumar (1983) and Kakar (1985). In a preliminary study of 2500 groundwater samples (2000 from dug wells and 500 from deep tube wells) from different parts of India, the incidence of high nitrate was conjectured to be due to pollution of some sort, either from human or animal sources or from irrigation return flows from agricultural fields dressed with chemical fertilizers (Handa, 1975). He also observed that nitrate in deep waters was only 1-2 mg/l whereas in shallow water it was up to 100 mg/l in humid areas and up to 1000 mg/l in arid and semi-arid regions.

Table 2. Statewise consumption of nitrogenous fertilizers in India compared with NO_3 in groundwaters and the stage of groundwater development

	Nitrogenous fertilizer consumption (kg/ha/yr)	Average NO_3 in groundwater (mg/l)	Maximum NO_3 reported in groundwater (mg/l)	Present groundwater development (1990) (% of total available)
State	I	II	III	IV
Jammu & Kashmir	1.40	7.90	275	1.24
Northeastern States	0.92 $	6.80	45	Negligible
Himachal Pradesh	4.20	8.60	177	21.81
Punjab	182.33	55.10	567	99.38
Haryana#	91.06	99.50	1800	70.15
Uttar Pradesh	52.56	22.60	634	36.00
Bihar	23.60	21.00	350	
West Bengal	43.00	14.20	480	16.54
Gujarat	22.16	<50.00	410'	30.80
Madhya Pradesh	8.40	30.20	473	12.46
Maharashtra*	10.59	>45.00*	?	22.04
Orissa (State)	8.53	14.80	?	4.80
Orissa(Ganjam District)**	22.60	213.00**	800	?
Andhra Pradesh	14.60	13.20	208	20.10
Karnataka***	20.43	>45.00***	200	33.93
Tamil Nadu	30.70	26.00	1030	47.00

Data sources:

(I) Author's calculations based on fertilizer consumption data of the Government of India (1991).
(II) & (III) After Handa (1986).
(IV) CGWB's special issue on groundwater statistics 1991.
* NO_3 content after Bulusu & Pande (1990), represents 23% of samples from Nagpur and Satara Districts.
** NO_3 content after Mehta *et al.* (1990), average values represent 22% of samples.
*** Represents 47% of samples from Gulbarga and Bangalore districts (based on Bulusu & Pande, 1990; Tamta *et al.*, 1992).
$ Average of seven northeastern states.
\# The consumption of fertilizers in Haryana has increased to 118.0 kg/ha in 1994-95 from a meagre 2.94 kg/ha in 1966-67 (Ahlawat *et al.*, 1996).

A rapid reconnaissance of nitrate in shallow groundwaters was done by CGWB (Handa, 1986). This study reported average concentrations of NO_3 in well waters (mostly from dug wells) from several states, namely Andhra Pradesh, Bihar, Delhi, Gujarat, Himachal Pradesh, Jammu and Kashmir, Madhya Pradesh, North Eastern India, Orissa, Punjab, Tamil Nadu, Uttar Pradesh and West Bengal with the exclusion of Goa, Karnataka, Kerala, Maharashtra and Rajasthan (Table 2, Columns II and III). Although the author mentioned towns and districts where concentrations in excess of 100 mg/l NO_3 were found, the number of samples analysed statewise was not indicated, and there was no discussion as to possible sources of pollution in different regions, i.e. whether from point-source emissions (urban sewage/industrial effluents) or non-point sources.

Subsequently, a study undertaken by NEERI reported nitrate in groundwaters from selected districts in 17 states of India including Rajasthan and Karnataka but excluding north-eastern states, Goa and Kerala (Bulusu and Pande, 1990). The authors reported high nitrate concentrations (above the WHO limit of 45 mg/l NO_3 for potable water) in 1290 out of 4696 samples (i.e. in 27% of samples) without referring to specifically either urban/industrial or agricultural sources. However, high nitrate levelss reported in 73% of samples from Nagpur metropolitan city, for instance, may be due to urban sewage and industrial sources. High nitrate content (45 to more than 600 mg/l) in shallow and deep tube wells due to seepage from industrial effluents and urban sewage has also been reported from around Jodhpur city in Rajasthan (Mathur and Ranganathan, 1990) and Lucknow city in Uttar Pradesh (Singh *et al.*, 1991). In Bangalore district (Karnataka) 18% of samples, representing 1475 km^2 were found to contain more than 50 mg/l nitrate, the maximum being 200 mg/l (Tamta *et al.*, 1992). Diffused ingress of seepages of nitrogen-bearing effluents were the cause in all these cases.

Recently Khanna *et al.* (1994) reported 23 analyses of groundwater samples obtained from shallow dug wells located in Sirsa, Hissar, Mahendragarh, Bhiwani and Rohtak districts. It was observed that 13 (57%) of samples contained more than 45 mg/l of NO_3. In nine samples NO_3 was above 100 mg/l, while it was above 300 mg/l in six samples, the maximum being 1030 mg/l in one sample. Although the source of nitrate was not discussed, in these rural districts the high nitrate could be due to diffuse pollution related to agricultural return waters.

In some 58 samples of water collected from top soil, gravel and laterite horizons (2 to 14 m depth) in Jhansi district of Central India, high nitrate levels (50 to 108 mg/l) were found in seven samples (12.5%) (Chandu *et al.*, 1995). Bhide (1990) has studied the nitrate pollution potential of Indian urban solid waste capable of affecting water-table aquifers by leaching. This would also essentially be diffuse pollution, though not agricultural.

DIFFUSE AGRICULTURAL SOURCES AND NITRATE POLLUTION

The study by CGWB (Mehta *et al.*, 1990), attributing high nitrate levels in dug-well waters of Ganjam district (Orissa) to high N-fertilizer use, was the first to directly implicate agricultural diffuse pollution. Gupta (1992) also seems to conjecture that high nitrate levels in groundwater of Udaipur district (Rajasthan) could be due to a five-fold increase in the use of N-fertilizers in the district during the decade 1979-89, thus blaming agricultural diffuse pollution. In the study by NEERI (Bulusu and Pande, 1990), high nitrate concentrations in 41% of samples from villages around Nagpur metropolitan city, and 49% of samples (261 out of 529) from Gulberga district (Karnataka), might also reflect contributions from agricultural sources. Therefore, in the following sections, this author has attempted to correlate the available information on nitrate levels in groundwaters with the increasing application of nitrogenous fertilizers, coupled with high groundwater use in some states/regions of India, which would be indicative of the role of diffuse agricultural pollution.

N-FERTILIZER USE AND POLLUTION HAZARD IN INDIA

The consumption of nitrogenous fertilizer in the country increased seven-fold during the decade 1960-61 to 1970-71 from a meagre 210,000 tonnes to 1,487,000 tonnes. It then increased six-fold in the following two decades reaching 8,579,000 tonnes in 1991-92 (Fig. 1). The present consumption of nitrogenous fertilizer (N) in India is 62% of the total (N + P + K) fertilizers.

In Punjab and Haryana, the granaries of India, average annual fertilizer consumption had attained a level of 162.33 kg/ha and 91.06 kg/ha, respectively, in 1991 compared to much lower consumption in other states (Table 2, Column I).

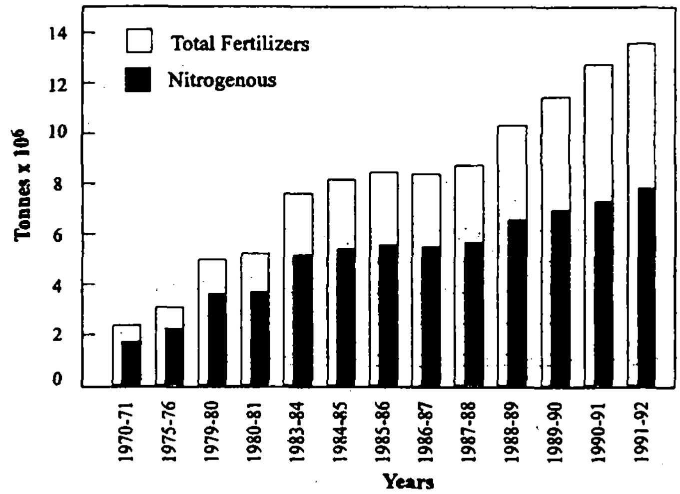

Figure 1. Consumption of nitrogenous (N) and total (N + P + K) chemical fertilizers in India over the past two decades.

An appraisal of the nitrate pollution potential of a region from non-point source application of nitrogenous fertilizers can be made from the consumption per unit area and water use for irrigation. Excess nutrients in irrigation return flow, if not properly drained-off, gradually infiltrate into aquifers. The plains of Haryana and Punjab constitute two principal states that have come under intensive agriculture.

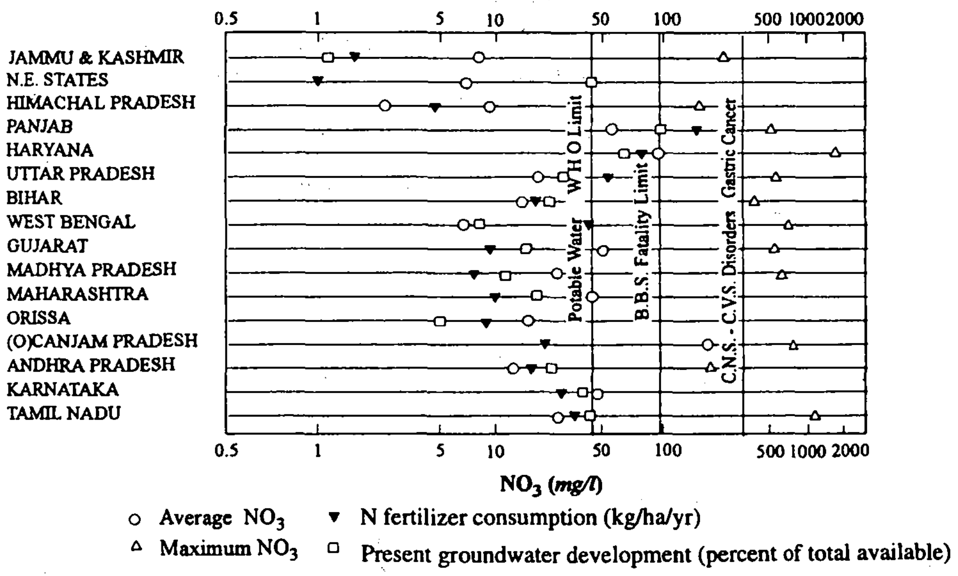

Figure 2. Groundwater nitrate levels in different states of India and health hazard limits represented with N fertilizer consumption and the stage of groundwater development.

The nitrogenous fertilizer consumption estimated by the author is maximum in these two states (Table 2, Column I). This correlates well with the high average nitrate levels in shallow groundwater of these states (Table 2, Column II). Whereas the average NO_3 content of the shallow groundwater in Punjab is above the upper limit recommended by WHO for potable water, the same in Haryana is close to the BBS fatality limit (Fig. 2). The figures in Column I of Table 2 are calculated on total surface areas of states and should not therefore be taken as the "average fertilizer dose" which will be still higher, as in each state some land is under forest cover and other non-agricultural use. Also, fertilizer consumption of a given district in a state may vary considerably from the average value for that district.

The maximum nitrate levels reported in some groundwater samples from Haryana, Punjab and U.P. in the north, Tamil Nadu and Karnataka in the south, Orissa (Ganjam district) and Bihar in the east, and Gujarat in the west, also parallels high average nitrate and high N fertilizer consumption. As would be expected, the average nitrate in groundwater modulates simultaneously with the consumption of N fertilizer and groundwater use. In the case of West Bengal, a low groundwater application level is reflected in a low average nitrate level in spite of high N fertilizer use. Better correlations may be obtained if more data are available at district level.

Agricultural diffuse pollution also depends on geology. Obviously irrigation without artificial drainage, in the poorly drained flat plains of Punjab and Haryana, comprising a thick pile of unconsolidated and permeable Late Quaternary-Holocene alluvial sediments (Lunkad, 1988), increases the nitrate pollution hazard compared to that in the freely drained regions of north and north-east India and the peninsular plateau of the south. Agriculture is not intensive in northern and north-eastern states as reflected in the meagre consumption of nitrogenous fertilizers and negligible groundwater use. Therefore, the nitrate pollution risk for groundwater is also minimum in these states (average NO_3 content below 10 mg/l). In the peninsular states the consumption of nitrogenous fertilizers and groundwater use are intermediate between those of northern and north-eastern states and the Haryana-Punjab plains. However the rising nitrate level in the groundwaters of this region should be viewed with concern for the future. Table 3 and Fig. 3 classify the states of India from the point of view of the potential hazard of nitrate pollution of groundwaters due to diffuse agricultural sources.

Figure 3. NO_3 diffuse pollution risk zones of India. 1: safe zone; 2: little or no risk; 3: increasing risk; 4: maximum hazard; ?: no or little data.

Table 3. Risk of groundwater nitrate pollution in different parts of India

Risk zone	Average N-fertilizer consumption (kg/ha)	Average NO_3 in groundwater (mg/l)	Groundwater development (% of total available)	Region (states)
1 (Little or no risk)	2	6 to 8	< 2 (negligible)	J and K, Northeastern States
2 (Low risk)	4 to 11	8 to 45	5 to 22	Himachal, MP, Orissa, Maharashtra
3 (Moderate risk)	14 to 53	13 to 50	16 to 4	UP, Bihar, W. Bengal, AP, Gujarat, Karnataka, Tamil Nadu
4 (High risk)	118 to 163	55 to 100	70 to 100	Punjab, Haryana

SUGGESTED MEASURES FOR PREVENTION OF DIFFUSE AGRICULTURAL NITRATE POLLUTION

Making the estimation of nitrate-nitrogen, as an indication of diffuse agricultural pollution, compulsory in routine chemical analysis of water. Nitrate concentration is often not reported in the current analyses.

Depth monitoring of descending nitrate plumes in aquifers either by estimation in existing wells penetrating different depths, or in test-bores especially driven for this purpose in affected and potential areas of pollution. This should be done quickly in the regions where intensive agriculture is practised.

Mandatory preparation of aquifer maps. Such maps should depict in plan and cross-section the depth and areal extent of the aquifers, distribution of shallow and deep wells through them, annual fluctuation (draft and recharge) of the water table or piezometric surface, storage potential and major quality indices such as EC, nitrate, phosphate, coliform index, etc. Sewage networks and industrial effluent disposal points in relation to surface-water bodies (rivers/reservoirs) should be superimposed on these maps. Information depicted on such maps should be updated regularly.

Introduction of legislation by the state governments restricting nitrogenous application to normal agriculture to a fertilization rate of about 120 kg N/ha per year. Splitting of fertilizer doses can also improve crop yield and reduce pollution of groundwater. Studies have shown that a single application may result in about 25-70 % losses to groundwater. One third or one half the application at the time of planting and another during rapid growth may result in high yields and reduce N losses to groundwater (Baien and Rykbost, 1976).

Spreading of cattle manure and liquid sludge should be banned during the time when aquifers are replenished by rains. A manuring season should be prescribed for main crops depending on monsoon and crop patterns in different states.

Enhancing the use of biofertilizers and green manures and encouraging research to improve their quality and efficiency.

Maintaining crop-cover during the required season and changing the crop patterns.

Encouraging forestry as an additional measure to protect groundwater resources in sensitive areas of nitrate pollution in Haryana and Punjab.

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