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Non-agricultural sources of groundwater nitrate: a review and case study

Fernando T. Wakida[a,*], David N. Lerner[b]

[a]*Facultad de Ciencias Químicas e Ingeniería, Universidad Autónoma de Baja California, Calzada Universidad, Mesa de Otay, Tijuana, Baja California, México*

[b]*Groundwater Protection and Restoration Group, Civil & Structural Engineering, University of Sheffield, Mappin St, Sheffield, S1 3JD, UK*

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Abstract

Nitrate is often seen as an agricultural pollutant of groundwater and so is expected to be at higher concentrations in the groundwaters surrounding a city than in those beneath it. However the difference between rural and urban nitrate concentrations is often small, due to the non-agricultural sources of nitrogen that are concentrated in cities. This paper illustrates the source and significance of non-agricultural nitrogen for groundwater and presents a case study of nitrate loading in the city of Nottingham. Major sources of nitrogen in urban aquifers are related to wastewater disposal (on-site systems and leaky sewers), solid waste disposal (landfills and waste tips). The major sources of nitrogen in the Nottingham area are mains leakage and contaminated land with approximately 38% each of a total load of $21\,kg\,N\,ha^{-1}\,year^{-1}$.

Keywords: Urban; Groundwater; Nitrate; Nottingham; Contamination; Leakage

1. Introduction

Nitrate is a compound of nitrogen that occurs naturally in moderate concentrations in many environments. Baseline concentrations of nitrate in groundwater beneath natural grassland in temperate regions are typically below $2\,mg\,l^{-1}$ (Foster et al., 1982). Because it is very soluble, it is the most usable form of nitrogen for plants. Nitrate is a common surface water and groundwater contaminant that can cause health problems in infants and animals, as well as the eutrophication of water bodies (Fennesy and Cronk, 1997).

Nitrate has been linked to agricultural activities due to the use of fertilizers. However, there are other nitrate sources related to urban development that can increase nitrate concentrations in groundwater. Studies in the last few years have found that nitrate concentrations in some urban aquifers are similar or even higher to those in their surrounding agricultural areas (Ford and Tellam, 1994; Lerner et al., 1999). The objective of this paper is to illustrate the sources and significance of non-agricultural sources of nitrate in groundwater.

2. Non-agricultural sources of nitrogen in groundwater

The wide range of pollutant sources and the complexity of recharge in urban areas make the estimation of

*Corresponding author. PMB # 184, 710 East San Ysidro Blvd "A", San Ysidro, CA 92173, USA. Tel.: +52 664 6821033; fax: +52 664 6822790.

E-mail addresses: fwakida@uabc.mx (F.T. Wakida), d.n.lerner@sheffield.ac.uk (D.N. Lerner).

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pollutant load a difficult task in these areas. This is also true for the various forms of nitrogen (oxidized and reduced forms) that are present in many possible recharge sources of urban aquifers. They include sewage and mains leakage, septic tanks, industrial spillages, contaminated land, landfills, river or channel infiltration, fertilizers used in gardens, house building, storm water and direct recharge. Because of the number of nitrogen sources in urban areas, it is not surprising to find elevated nitrogen concentrations in urban aquifers. The sources of nitrogen in the urban environment are a mixture of point sources (e.g. landfills and coal gasification works), multipoint sources (e.g. soakaways and leaky sewers) and diffuse sources (atmospheric deposition, house building and recreation areas). An overview of these sources is presented in the following sections and Table 1 reviews some published cases.

2.1. Leakage from water supply and disposal networks

Leakage from sewerage and water supply networks provides the highest percentage of water recharge to aquifers underlying many cities through out the world (Yang et al., 1999; Lerner, 1986). Water mains and sewers leak because of improper installation or deterioration through age, subsidence or earthquakes. Sewage leakage occurs when sewers are situated above the water table. Few studies have attempted to quantify the recharge and pollutant load from leaky sewers. Published examples of leaky sewers studies are mainly from Germany. In Hanover, 5–8 $Mm^3 year^{-1}$ of sewage is entering to the aquifer (Mull et al., 1992). It is estimated that more than 100 $Mm^3 year^{-1}$ of wastewater percolates from damaged sewers to subsurface in Germany (Eiswirth et al., 2000). On the other hand, mains leakage is almost an inevitable fact for water supply companies and losses of 20% are considered routine in the United Kingdom (UK). For Liverpool, it was calculated a leakage of 36.5 $Mm^3 year^{-1}$ equivalent to a recharge of 180 $mm year^{-1}$ (Price and Reed, 1989) and in the city of Amman, Jordan the leakage from water-supply system is approximately 24 $Mm^3 year^{-1}$ (Salameh et al., 2003). This review has shown leaky sewers and mains have a major impact on groundwater quality. Leaky sewers contribute with a wide range of pollutants such as bacteria or organic and inorganic compounds. On the other hand, water supply is often of good quality. Nitrogen concentrations are likely to be under the drinking water limit of 10 $mg l^{-1}$ and so leakage can be beneficial for pollutant dilution.

2.2. On-site sewage disposal

On-site sewage disposal encompasses cesspools, septic tanks and pit latrines. It is estimated that in the United States approximately one third of its sewage is disposed of by septic tanks (Harman et al., 1996). Septic tanks are also a common practice in other countries such as Canada and Australia, as well as in developing countries (Harman et al., 1996; Whelan, 1988). In Sana'a, Yemen, it is estimated that 80% of the urban recharge is wastewater from cesspits (12.5 Mm^3 in 1993) (Alderwish and Dottridge, 1999) and in the city of Amman, Jordan, 8 $Mm^3 year^{-1}$ from cesspool leakages are recharging the aquifer (Salameh et al., 2003). Conversely, in the UK only 5% of the population is not served by mains sewerage and most of these are served by septic tank systems (Payne and Butler, 1993).

The concentration of total nitrogen in effluents from a typical septic tank system ranges from 25 to 60 $mg l^{-1}$, with ammonia making up the vast majority of this total, 20–55 $mg l^{-1}$ as ammonia and less than 1 $mg l^{-1}$ as nitrate (Canter, 1997). Ammonium ions in the effluents may be oxidized to nitrate which can be transported in the subsoil beneath the septic tank absorption field and subsequently to groundwater.

The density of systems is the most important factor in groundwater contamination by septic tanks. A minimum lot size of 0.4 to 0.6 ha is needed to insure against groundwater contamination (Bicki and Brown, 1991). Other factors that can contribute to the hazard of groundwater pollution by septic tanks are improper design, poor maintenance and depth of the water table (Yates, 1985).

A greater threat to groundwater quality is the use of pit latrines because of the discharge of waste without pre-treatment. Human excreta contains about 5 $kg N year^{-1}$ per capita (Lewis et al., 1980). Therefore, high population density plays an important role in the impact of this waste disposal method.

2.3. Animal waste

Animals are, or have been a common feature in many cities in the world. Cats, dogs and less commonly horses and urban wild animals are inhabitants in many cities. In the suburbs of cities in developing countries, it is common for people to keep some farm animals for consumption or for commercialization. For example, there are about 23,000 dairy cows in the urban and peri-urban areas of Addis-Abeba, Ethiopia (Bonnet and Duteurtre, 1999) and approximately 60,000 cows in the metropolitan zone of Mexico City (Lozada et al., 2000). Somasudaran et al. (1993) suggested the substantial number of oxen, cows and buffaloes in Madras, India, is one of the sources of the high nitrate concentration in shallow groundwater. Excreta, dung and urine produced by animals constitute a potential source of contaminants such as nitrate, potassium and bacteria. They can enter groundwater by way of storm water channels or river, recharge basin or direct recharge.

Table 1
Summary of some published research on non-agricultural sources of groundwater nitrogen

Source	Loading ($kg\,N\,ha^{-1}\,year^{-1}$)	Effluent concentration ($mg\,N\,l^{-1}$)		Concentration in nearby groundwater ($mg\,N\,l^{-1}$)		Location
		NH_4	NO_3	NH_4	NO_3	
Leakage from water supply and disposal network						
Leaky sewers	123	55	2		4–10	Rastatt, Germany (Eiswirth and Höltze, 1997)
	19[a]				5	Munich, Germany (Merkel et al., 1988)
	3	30			shallow: 10 deep: 12	Nottingham, UK (Barrett et al., 1999; Yang et al., 1999)
Leaky mains	8.3		5.6			Nottingham, UK (Barrett et al., 1999; Yang et al., 1999)
On site sewage disposal						
Septic tanks	100[b]		25		10–30	Merida, Mexico (BGS, 1995)
	47.5		68			New England, USA (Gold et al., 1990)
Contaminated land						
Landfill	3300–4400	1000–1350		6.4–67.5	0.04–0.2	Greece (Fatta et al., 1999)
	290–5700[c]	90–305	2–2.5	0–40	0–1.4	Zagreb, Croatia (Mikac et al., 1998; DoE, 1995)
		270–672	<0.1–7	17–35	<0.1–1	USA (Murray et al., 1981)
Gasworks	54000	9956		285	107	Mansfield, UK (Davison and Lerner, 1998)
River–aquifer interaction					Increase from 1.8 to 5 in 8 days	Arkansas, USA (Sophocleous et al., 1988)
Atmospheric deposition	5–35					UK (DoE, 1994)
Urban fertilizer						
	210	0.02–0.59	0.85–5.37			Perth, Australia (Sharma et al., 1996)
	1–55		0.1–18.9			USA (Snyder et al., 1984)
Storm water						
Highways and roads	3.2–4.6		1.4–3.3		1–3	USA and Europe (Luker and Montague, 1994)
	0.7–8.7		0.37–1.07			Texas, USA (Barrett et al., 1998)
Urban		0.1–3.5	0.0–2.70			USA and Europe (Makepeace et al., 1995)
Airfield deicing	1245[d]	Conc. in stream 143				Newcastle, UK (Turnbull and Bevan, 1995)
House building	59		48–303			Nottingham (Wakida, 2002)

[a]Assuming a concentration of $80\,mg\,N\,l^{-1}$, similar to the concentration of the sewage of Rastatt, Germany (Eiswirth and Höltze, 1997).

[b]The loading was calculated using an estimated recharge and nitrate concentration of $400\,mm\,year^{-1}$ and 25 mg NO_3–N Loading rates based on a observed loss of $9.5\,kg\,year^{-1}$ from a three-person home and a density of five homes ha^{-1}.

[c]All the data, except the loading values, are from (BGS, 1995). The loading values were calculated using the minimum and maximum leachate production estimated in (Gold et al., 1990) (3200 and $18{,}592\,m^3\,year^{-1}$) and the mean NH_4 concentration in the leachate. It must be stressed that these values can be very different to those observed due to the different conditions used (rainfall, waste density, landfill age, etc).

[d]Application rate in the winter 1991/92. Nowadays urea based de-icer is not used in commercial airports in the UK.

An estimated horse dung production between 100,000 and 200,000 tones year^{-1} in 1910 in the urban Birmingham aquifer (Fernandes and Tellam, 2001). Although dung production was considerable, it would be probable that the waste was concentrated in particular zones, such as stables and heavily used roads, and part of the dung would have been collected for fertilizer use. According to the authors it is unlikely that horse dung has been a dominant contributor of N concentration in the aquifer, but local severe pollution close to stables appeared to be probable. In the other hand, a study conducted in Long Island, USA, estimated a small load of 9 kg N ha^{-1} year^{-1} from cats and dogs in a housing development (Flipse et al., 1984). Based on the above cases, animal wastes can be a significant source of nitrate to urban groundwater in cities of developing countries, but not in developed countries.

2.4. Contaminated land

Contaminated land, such as abandoned landfills, gasworks sites or abandoned industrial sites, contribute a significant quantity of nitrogen to groundwater. Landfills are considered a major source of pollutants and their impacts on groundwater quality have been extensively reported in the last four decades (Zanoni, 1972; Walls, 1975; MacFarlane et al., 1983; Reinhard et al., 1984; Albaiges et al., 1986; Flyammar, 1995). In the UK, it is estimated that there are at least 5000 closed landfills (De Hénaut and Harris, 1996). These are mostly now within urban areas because, even if they were originally in the outskirts, the cities have expanded to engulf them. Landfill leachate has a high content of N, organic compounds and heavy metals. Most of the N found in leachate is in the form of ammoniacal nitrogen due to the anaerobic conditions prevailing in landfills. Typical concentrations of N are ammonium, 0–1250 mg l^{-1}; nitrate, 0–9.8 mg l^{-1} and nitrite, 1.5 mg l^{-1} (Arigala et al., 1995). Landfills in many developing countries are usually no more than open dumps. Moreover, the unplanned growth of cities and the lack of rubbish collection service have developed dumpsites within the built areas. For example in Lagos, Nigeria, maximum N concentrations in groundwater samples in the vicinity of dumpsites within built area were between 84 and 124 mg l^{-1} (Adelana et al., 2003a).

Before the discovery of natural gas in the North Sea in the 1960s, coal was gasified for heating and lighting purposes in many northern European countries. Many towns had their own gasworks. Waste from these plants was disposed on the ground creating serious groundwater and soil pollution by organic and inorganic compounds. Ammonia concentration in leachates has been found in the range of 50–1000 mg l^{-1} (Davison, 1998). There are approximately 10,000 coal gasification related sites in the UK (De Hénaut and Harris, 1996), and as in the case of landfills, most of them are in urban areas.

An example of contaminated land arising from industrial use are the military facilities involved in the production and demolition of explosives, propellant and pyrotechnics. They are potential sources of numerous contaminants of groundwater including nitrate. Nitrate is formed by the biodegradation and combustion of explosives such as cyclotrimethylene-trinitramine ($C_6H_6N_6O_6$) commonly known as RDX, hexogen or cylonite. Several sites with explosives contamination have been identified in the United States and concentrations of N in groundwater in these sites range from 20 to over 200 mg l^{-1} (ITRC, 2000).

The high concentrations of N and other inorganic and organic compounds in the leachate from abandoned landfills, gasworks sites and some abandoned industrial sites made them significant sources of pollution to aquifers. They are likely to persist for years given the historical numbers of unlined landfills and unremediated sites.

2.5. Industry

Current industry is another potential source of nitrogen to groundwater. Nitrogen compounds are used extensively in industrial processes. Some examples of industrial uses are plastic and metal treatments, raw materials for the textile industry, particleboard and plywood, household cleaning and the pharmaceutical industry (Potash Corporation of Saskatchewan, 2002). The predominant nitrogen compounds used in industry are ammonia, nitric acid, urea and ammonium nitrate (Potash Corporation of Saskatchewan, 2002). Nitrate contamination may result from inadequate handling, disposal or use of these compounds. For example, a high proportion of the sulfate in the Birmingham aquifer is from industrial spillages, mainly of sulfuric acid (Hughes et al., 1999). Therefore, it is expected that nitrogen compounds (e.g. nitric acid) from spillages will have a significant impact on groundwater quality in industrial zones.

2.6. River and aquifer interaction

Aquifer contamination by river or unlined canal carrying highly polluted water has been reported elsewhere (BGS and MoPH, 1994; Kacaroglu and Gunay, 1997). There are two factors that lead to aquifer contamination by a river; firstly, the river has received a high proportion of raw or treated wastewater, and secondly the river is infiltrating water to an aquifer. Usually, the second factor occurs when pumping stations are near the river. Most of the published reports on aquifer pollution by rivers are for karstic aquifers (McConnell and Hacke, 1993; Plummer et al., 1998;

Personé et al., 1998; Crandall et al., 1999), when they are connected hydrologically. Other examples are in alluvial aquifers (mostly from Europe) which are used for riverbank infiltration (Schwarzenbach et al., 1993; Von Gunten et al., 1986; Doussan et al., 1997). Denitrification and mixing (Doussan et al., 1998; Grischek et al., 1998) can reduce nitrate concentrations during river water infiltration and streambed sediment can act as a barrier to pollution as in the Thames Basin (Younger et al., 1993). River–aquifer interaction is not particularly an urban issue, except that river pollution is usually produced by urban sources such as wastewater and industrial effluents.

2.7. Atmospheric deposition

The emission of nitrogen to the atmosphere can be in its oxidized or reduced forms. The oxidized forms are mostly generated by car engines and industry (NO_x) and the reduced species (ammonia) are mainly from agriculture and intensive feedstock rearing. These forms can later be deposited and be carried in roof and storm water runoff. Atmospheric deposition can contribute on average some 40–45% of nutrients and heavy metals associated with storm water discharged from urban catchments (Ellis, 1986). The Department of the Environment has quantified emissions and depositions of nitrogen in the UK. The highest depositions range between 10 and 20 kg N ha^{-1} year^{-1} and are in the zones with the highest rainfall (DOE, 1994).

2.8. Urban fertilizer use (gardens, recreational grassland and domestic horticulture)

Nitrate leaching from fertilizers applied to lawns, gardens and urban plots for cultivation of vegetables is likely to be a significant source of nitrogen to groundwater. The use of fertilizers in gardens, residential lawns and recreational grassland such as golf courses has been identified as one of the sources of nitrate in urban aquifers (Sharma et al., 1996; Katz et al., 1980; Wong et al., 1998). In a study carried out in a rural community in Germany, the researchers found that home gardens with a cover of only 3.5% of the total study area were responsible for 27% of the total amount of nitrogen leached (Kliebsch et al., 1998). The leaching of nitrate from fertilizer applied to turfgrass depends highly on soil texture, N source, rate and timing, and irrigation/rainfall (Petrovic, 1990). The worst case scenario for nitrate leaching is an application of a soluble N source at a rate higher than the recommended rate, to a sandy site that is over irrigated. However, nitrate leaching from turfgrass can be reduced with good management practices (Petrovic, 1989).

Urban food production is increasing around the world, wedges of land in many cities are used for vegetable gardens. For example, New York has more than 1000 community gardens and Berlin some 80,000 (Anonymous, 1996). Growing vegetables in cities is a very common activity in many developing countries. In Lusaka, the capital of Zambia it is estimated that nearly 60% of low-income households are cultivating food gardens (Sanyal, 1985). Meanwhile, in Harare, Zimbabwe, 16% of the city area is dedicated to urban agriculture (Anonymous, 2004).

Pionke et al. (1990) determined the impact of irrigated horticulture on groundwater. They found high concentrations of N in drainage below root zone (71–209 mg l^{-1}) and 9–80 mg l^{-1} in shallow groundwater. Thompson et al. (2002) found a mean value of soil mineral N of 527 kg ha^{-1} at a depth of 60 cm in horticultural greenhouses, showing the high pollution potential on the underlying aquifer. Nitrogen load from domestic horticulture through out the world is likely to be very variable. It depends on management practices and the area used for this activity. Based on the literature reviewed, it is likely that fertilizer used in urban agriculture may be a significant N source to groundwater in some fast growing cities in developing countries.

2.9. Storm water

It is well known that highway and urban area drainage can contain a wide range of pollutants. Nitrogen compounds in highway drainage can originate from a variety of sources. These sources include atmospheric deposition, decaying organic matter, dispersion of fertilizers from adjacent agricultural land, and less frequently, accidental spillages of material containing nitrogen, e.g. fertilizers, slurry, sewage sludge, etc (Bellinger et al., 1982). Although groundwater pollution cases by road pollutants have been reported (Saleem, 1977; Howard and Beck, 1993), storm water is not a major source of N to groundwater because of its low N concentrations, as is shown in Table 1.

2.10. House building

Nitrate leaching due to house building is potentially equivalent to that due to ploughing of pasture, which has been identified as a major source of nitrate in groundwater beneath agricultural land (Whitmore et al., 1992). Soil disturbance during house building increases the soil aeration and the mixing of nitrogen and carbon sources with soil organisms. Soil microbial activity in favorable conditions leads to an accumulation of nitrate due to mineralization and nitrification processes. Nitrate will be leached by drainage of excess water if it is not taken up by plants (McLenaghen et al., 1996). A recent study has shown that house building activities lead to leaching of nitrate to groundwater. The average

Table 2
Nitrogen sources identified in selected urban areas

Continent country city	Conc. in groundwater (mg N l^{-1})	On site sewage disposal	Leaky mains	Leaky sewers	Contaminated land (landfills waste disposal)	Industry	River or channel aquifer interaction	House building	Urban fertilizers use	Reference
America										
USA										
Minnesota	1–21	*								Woodwart et al. (1961)
Long Island, NY	0.1–3 (a)								*	Flipse et al. (1984)
Mexico										
Mexico City		*		*	*	*	*			Mazari and Mackay (1993)
Tijuana, Baja California	0.5–50	*								Guzman Garcia (1998)
Merida, Yucatan	10–30	*								BGS (1995)
Bolivia										
Santa Cruz	Shallow: 10–40 Deep: 5–25	*								BGS (1994)
Argentina										
Azul	9 (a)	*								Usunoff and Varni (1995)
Bermuda	Unsewered: 25–35 Sewered: 5–10	*		*						Thomson and Foster (1986)
Europe										
France										
Narbonne	1–34	*				*				Razack et al. (1988)
United Kingdom										
Birmingham	2–21			*	*	*			*	Ford and Tellam (1994)

Nottingham	Shallow: 10		*	*	*	*		*	*	Barrett et al. (1999); Yang et al. (1999)
	Deep: 12 (a)									
Germany										
Hannover	1–17			*	*				*	Mull et al. (1992)
Turkey										
Eskisehir	3–23 (a)	*					*			Kacaroglu and Gunay (1997)
Asia										
India										
Lucknow	10–130	*								Sahgal et al. (1989)
Madras	3–226	*		*			*			Somasundaram et al. (1993)
Karnataka	9–112	*								Ramaraju et al. (1999)
Thailand										
Hat Yai	2–22						*			BGS (1994)
South Korea										
Taejon	2–9 (a)	*		*	*					Jeon (2001)
Israel										
Israel	6–25	*								Burg and Heaton (1998)
Africa										
Senegal										
Dakar	24–70 (a)	*			*					Cisse et al. (2000)
Zambia										
Lusaka	15–18	*			*	*				Nyambe and Maseka (2000)
Nigeria										
Lagos	0–64 16 (a)	*			*	*				Adelana et al. (2003b)

(a) average

potential nitrate loss from sites under construction was estimated as 59 kg N ha^{-1} year^{-1} (Wakida, 2002). The most important factors affecting the potential of nitrate leaching from house building include previous land use, quantity of total nitrogen in the remnants after the stripping of topsoil and date of commencement of works. The first two factors are intimately related because previous land use determines greatly the soil conditions before and during house building. The date of commencement of work will determine the time of soil disturbance, thus the potential of mineralization and nitrification of soil organic nitrogen.

3. Source and significance of non-agricultural N to groundwater

Urban sources of N may have a high impact on groundwater because of the high concentration of potential sources in a smaller area than agricultural land. Table 2 reviews the identified sources of N for a variety of urban aquifers around the world. The examples in Tables 1 and 2 show that nitrogen in urban groundwater can be similar to or even higher than concentrations in equivalent rural areas. These examples show that leakage from underground networks is a common feature in urban areas. Leaky sewers appear to be a worldwide problem, but is very variable between urban areas. In Nottingham, sewage leakage is not a significant N source, while some German cities have higher N loads from sewage (Table 1).

Mains leakage plays an important role in the hydrogeology in some urban areas. It is a major source of urban recharge and loss rates of 20–25% are considered normal in the UK (Price and Reed, 1989). Nitrogen concentrations in water from mains is usually low (<10 mg l^{-1}), but the large volume of leakage from mains produces a large nitrogen load. For example, in Nottingham is estimated that around 55,000 kg year^{-1} are polluting the aquifer. However, as mentioned above, mains leakage is usually seen as beneficial because of its good quality provides dilution for pollutants. A reduction of the recharge by mains leakage may have detrimental effects on urban groundwater quality.

Effluent from septic tanks is a frequently cited source of high nitrate concentrations in urban groundwater. The issue of wastewater disposal is a more serious problem in cities of developing countries where, generally, there are many high dense populated and unsewered areas created by high rates of migration into cities. These areas are unplanned and located in the outskirts of the cities forming shanty towns where pit latrines or septic tanks are common. In some cities septic tanks and pit latrines are the only way to dispose of sewage, while groundwater is the main water drinking source. N concentrations in many groundwaters below unsewered cities are between 10 and 50 mg l^{-1}, but the impact of on-site disposal systems on groundwater will depend on many factors (Morris et al., 1994). But even if these areas or cities are served by sewerage, the problem is not solved because high proportion of wastewater is discharged to channels or rivers with little or no treatment. This can pollute the aquifer as in the example of Madras, India, where concentrations up to 1000 mg l^{-1} were found in groundwater samples. Infiltration of sewage from unlined channels and rivers is likely to be a more widespread problem than the literature review has shown, since many cases have not been reported.

Contaminated land is one of the most important sources of groundwater pollution. In developed countries, new landfills will not be a problem in the future because of the strict environmental control of landfill construction and operation. However, because of their large numbers and the high pollutant concentration associated with them, it is likely that landfills and former industrial sites will continue to be a major source of N to groundwater.

Non-agricultural sources of N are linked to other pollutants that threaten groundwater quality more seriously, such as bacteria and organic compounds in sewage, leachate from landfills, or phenol, BTEX and heavy metals from coal gasification plants. Therefore, urban groundwater can be seriously polluted. However, the pollution extent and nature will depend on many urban and natural attributes of the city such as geology, climate, industry types present, population density, and conditions and location of sewerage and mains network.

4. Case study: non-agricultural sources of N in the Nottingham area

4.1. Background

Nottingham in the UK has been the setting for a series of city-scale research projects on urban groundwater. These have reported on chemical and microbiological quality of groundwater (Barrett et al., 1999; Powell et al., 2003; Cronin et al., 2003), urban recharge (Yang et al., 1999), groundwater flow patterns (Trowsdale and Lerner, 2003; Taylor et al., 2003), and the risks of pollution from contaminated land (Tait et al., 2004). These data and models have enabled us to estimate the total nitrogen loading to groundwater, and its subdivision between sources.

Nottingham was chosen for the study because it has no significant superficial cover to complicate interpretation, and has rural areas nearby for comparison. In addition, the aquifer is mostly oxic, so N is generally as nitrate in groundwater, although it could be organic or ammoniacal N near to some sources before oxidation in the aquifer. The hydrogeology of Nottingham is described in previous publications (e.g. Yang et al., 1999; Trowsdale and Lerner, 2003), and is only briefly summarized here.

Groundwater is principally found in the Triassic Sherwood Sandstone Group. This is a fluviatile red-bed sequence which varies in thickness from zero in the west to over 150 m in the north. A typical regional hydraulic conductivity is a few m/d. Matrix porosity is about 25–35%, specific yield about 10–15%, and there is some fracturing. It is confined to the east and south by the Mercia Mudstone Group, and overlain in the valleys of the rivers Leen and Trent by alluvium. Regional groundwater flow is to the south and east, discharging into boreholes with high pumping rates and the two rivers. Pumping for public supply started in the late 19th century, and is continued by a network of one urban and six rural pumping stations, supplemented by surface water.

4.2. Methods

There were two major steps in the analysis. In the first, the total recharge and average concentration of N in recharge were estimated, as explained below; together these give the total load of N entering the aquifer. In the second step, the load of N was subdivided between the various sources, again as described below.

4.3. Total recharge and N loading

The land use and infrastructure of a city are too complex to be able to quantify recharges directly for each parcel of land, without excess effort. Hence this study focused on making areal averages of each kind of recharge. There are three principal sources of recharge for Nottingham and each is expected to vary in space and time. They are precipitation recharging through open spaces such as gardens and parks, leaking water mains, and leaking sewers. Our approach has been to simultaneously calibrate four flux balance models, thereby maximizing the information used and minimizing uncertainty in the outcome. The fluxes were groundwater and the three conservative chemical species, chloride, sulphate and total nitrogen. A groundwater flow model and three solute transport models were constructed and calibrated against groundwater level hydrographs and all available measurements of solute concentrations, including a few measurements from the 19th century. The area was divided into six zones, and the study period of 1850–1995 was divided into six different recharge and solute periods, based on analysis of the growth of the urban area. Time was further subdivided into 13 periods to include variations in pumping. The models used much smaller discretisations to solve the governing equations, with grid sizes down to 250 m and time steps of 1 year.

Full details of the methods and results have been published (Yang et al., 1999) and are only summarized here. The outcomes of the interlinked regional models for flow and solutes were (a) estimates of total recharge for each of the six spatial zones for each time period, (b) subdivision of total recharge between open space, leaking mains and leaking sewers, and (c) estimates of average N content in recharge for both rural and urban areas. Averaging the recharge estimates over the urban area for the most recent period (1958–1995) gives:

Total recharge	211 mm year^{-1},
Recharge from mains leakage	138 mm year^{-1},
Recharge from sewer leakage	9 mm year^{-1}.

The average N concentration in total recharge showed similar rising trends in rural and urban areas. In rural areas, N concentrations rose from 5 to 13 mg l^{-1} between the mid 19th century and the 1990s, while urban recharge concentrations rose from 3 to 17 mg l^{-1} over the same period (Yang et al., 1999). The latter is equivalent to a loading of 21 kg N ha^{-1} year^{-1}.

4.4. Components of the N loading

Five major sources of N were identified from a review of the city structure:

1. Leaking water mains,
2. Leaking sewers,
3. Releases of N during house building,
4. Leaching from soil in open land such as gardens and parks,
5. Contaminated land including landfills, industrial land and chemical spills.

The N loads associated with the water mains and sewers were relatively easy to estimate. Average concentrations in each source were available from modern measurements (5.6 and 30 mg l^{-1}, respectively), and, in the absence of any better information, were assumed to apply to the whole period of study. The hydraulic loadings from mains and sewers were estimated as part of the recharge exercise summarized above.

Releases from house building were estimated from a study of N profiles in the unsaturated zone at several building sites in and around Nottingham (Wakida, 2002). An average of 59 kg N ha^{-1} year^{-1} was calculated for the building sites. Maps of the city were used to estimate the areas of house building, and hence derive average loads over the city, for each period.

Leaching from parks and gardens could only be estimated by making three major assumptions. These were that the soil processes in open land have caused a constant concentration of N in recharge over the period, that this concentration is 3 mg l^{-1}, and that the proportion of open space in built-up areas has stayed

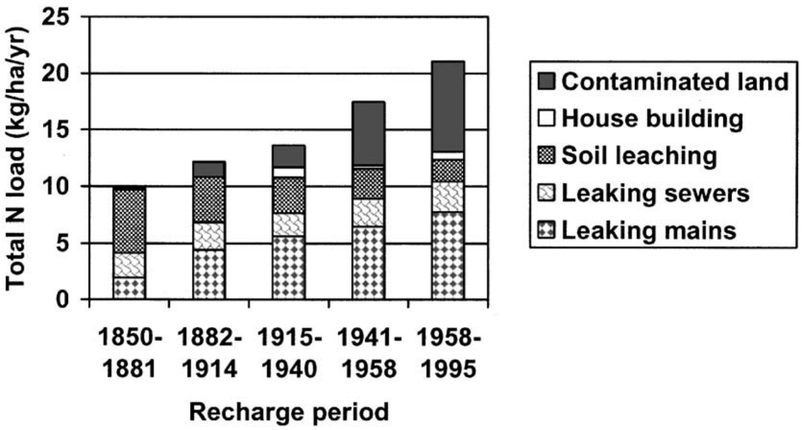

Fig. 1. The composition of nitrate load for different periods in the Nottingham urban area.

constant. The first assumption is possibly justified by the relatively infrequent use of fertilizers in UK gardens. The assumed concentration is the average concentration estimated for the earliest period of the study, and is similar or a little higher than concentrations observed under undisturbed grassland (Young and Gray, 1978). The final assumption on the proportion of open space is open to question, but may be justified because the city has grown by a series of developments of new housing around the perimeter, rather than by infilling.

4.5. Results

With estimates of N loading available for the first four sources, the loading from contaminated land was estimated by difference. The resulting estimates of loads for all five sources are shown in Fig. 1. They are loads for the unconfined aquifer in the current urban area of Nottingham and are averages over the three zones which are now urban. The uncertainties are clearly greatest for the early periods, for which few data are available, and for the contaminated land source which has been estimated by difference. Nevertheless, this is possibly the only city for which any estimates are available from integrated analysis of chemical and hydrogeological data.

The modern loads are distributed between leaking water mains 37% (7.7 kg N ha^{-1} $year^{-1}$), leaking sewers 13% (2.7 kg N ha^{-1} $year^{-1}$), soil leaching in open spaces 9% (1.9 kg N ha^{-1} $year^{-1}$), house building 3% (0.7 kg N ha^{-1} $year^{-1}$), and contaminated land 38% (8.0 kg N ha^{-1} $year^{-1}$). The highest N load from house building was in the period 1915–1940 during the period of major expansion in Nottingham. House building can have negative local impacts on groundwater quality, but will not be significant on the regional scale. Leaky sewers' contribution of N load are also not significant on the regional scale, but can produce serious groundwater pollution and public health problems due to the serious pollutant involved, such as disease causing organisms and industrial chemicals. The major sources of N in the Nottingham area are leaking water mains and leaching from contaminated land.

5. Conclusions

Differences between N concentrations in groundwater from aquifers underlying agricultural and urban areas are small, and sometimes N concentrations are higher in urban areas. The number of potential sources, the high density of these sources, and the concentrated nature of some of them such as landfill leachate or sewage, produce high N concentrations in groundwater.

This review has shown that the major sources of N in urban aquifers throughout the world are mostly related to wastewater disposal (on-site systems and leaky sewers) and solid waste disposal (landfills and waste tips). In developing countries, a sewage collector system and treatment in conjunction with an appropriate solid waste disposal system will reduce much of the problem. However, it will not eliminate the risk of groundwater pollution by leaking sewers.

Estimates of the total urban load of N and its components have been made for the Nottingham aquifer, mainly through the development of a set of four flux balance numerical models, for water, chloride, sulphate and total N. The total load in recent times is 21 kg N ha^{-1} $year^{-1}$, and the major components are mains leakage (37%) and contaminated land, including landfills, with 38%.

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