

# Diffuse pollution of groundwater in urban areas

David N. Lerner

Catchment Science Centre, University of Sheffield

## Abstract

The multiplicity of pollution sources in cities can be seen as equivalent to diffuse pollution, rather than a set of point and linear sources. Very little attention has been paid to the loads and impacts of such diffuse pollution. Sustainable urban hydrology must consider how to estimate such loads, in order to design better urban infrastructures to reduce the loads to acceptable levels. This paper presents two approaches to estimating loads, the component and the integrated approaches. These are illustrated with examples from Nottingham and Birmingham for nitrogen and industrial pollutants (MTBE and chlorinated solvents). The total N load was estimated to be  $2.1 \text{ g m}^{-2} \text{ y}^{-1}$ . The MTBE load estimate is  $1 \text{ g m}^{-2} \text{ y}^{-1}$ , although this is suspected to be too high as it is dependent on the biodegradation rate chosen. The chlorinated solvent load estimate ranges from 0.04 to  $0.1 \text{ g m}^{-2} \text{ y}^{-1}$ . All of these loads are high enough to cause significant pollution of urban groundwater.

## Introduction

Groundwater supplies 30% of UK public water supply, and a much higher proportion in Europe and other countries. It also supports all perennial rivers and their ecosystems by providing all of the dry weather flow (except for the sewerage discharges). Urban groundwater is part of this important resource, often with a close connection with surface waters, but is at risk of pollution from city landuses. There is a tendency not to use urban groundwater because of these risks. With the ever increasing pressure on water resources and the requirements for environmental flows in surface waters, we cannot afford ignore this resource, and so we need to get a better understanding of the risks of pollution.

The problems of point source pollution of groundwater are well documented, and investigations of contaminated sites form a major part of the work of the hydrogeological consulting community. However, in urban areas there are many potential sources of groundwater pollution, many of which are not explicitly known. They include:

- Petrol stations and fuel storage facilities in industrial premises and transport companies;
- Chemical works, both large scale manufacturing works and the many smaller scale warehouses formulating products for sale;
- Manufacturing plants for metal products, electronics, clothing, etc. which all use chemicals for degreasing, as solvents and during processing;
- Retail points where chemicals are used as solvents, such as dry cleaners, auto repair shops and printing works;
- Landfills and other waste disposal facilities;
- Contaminated land where the original polluting activity

is no long active but residual pollution is present and continues to flush into groundwater;

- Parks, gardens and other open spaces where atmospheric fall-out, fertiliser and pesticide applications, and other diffuse loads can be flushed to groundwater;
- Leaking sewers.

There are many small sites which can release pollutants to groundwater, and they usually do. This is because, on average, the management of chemicals on small sites is worse and less regulated than on sites owned by large, sometimes multinational, companies. Even though the sites are small, they are large in number. Acting together, these many point sources can be considered to be equivalent to diffuse pollution. To manage urban groundwater, it would be helpful to know the net effect of this multiplicity of sources. The objectives of this paper are to discuss how the diffuse pollution load from urban sources can be estimated, and to give some examples.

## Estimating diffuse pollution loads

There are two approaches to estimating N loadings to groundwater, the *component* and the *integrated* approaches, each with advantages and disadvantages. The component approach examines all the sources listed above, whereas the integrated approach considers only their overall effect.

### *Component approach*

The component approach is the more intuitive. Each of the possible sources of the pollutant load is considered separately, the necessary information gathered to estimate

that component of the load, and the components summed. The sources can be divided into three types, each with different hydraulic characteristics and so requiring different methods of estimation. The first is where the pollutants are soluble and spread over an area (e.g. contaminated land, parks and gardens) and so are carried to groundwater by diffuse recharge. The second type is where a point (e.g. soakaway) or linear (e.g. leaking sewer) hydraulic load carries dissolved pollutants. The third is where non-aqueous liquids (NAPLs) are spilt (e.g. dry cleaners, fuel leaks). NAPL in the unsaturated zone will be slowly dissolved and flushed to groundwater by recharge. NAPLs below the water table will be dissolved by passing groundwater. The total load ( $L$ ,  $\text{g m}^{-2} \text{y}^{-1}$ ) is given by:

$$L_T = \left( \sum_{\text{Land parcels}} R_i A_i C_i + \sum_{\text{Hydraulic loads}} Q_j C_j + \sum_{\text{NAPL sources}} L_{\text{NAPL},k} \right) / A \quad (1)$$

where  $R_i$ ,  $A_i$ ,  $C_i$  are the recharge rate ( $\text{m y}^{-1}$ ), area ( $\text{m}^2$ ) and concentration ( $\text{g m}^{-3}$ ) associated with land parcel  $i$ ;  $Q_j$ ,  $C_j$  are the water flow rate ( $\text{m}^3 \text{y}^{-1}$ ) and concentration ( $\text{g m}^{-3}$ ) associated with hydraulic load  $j$ ,  $L_{\text{NAPL}}$  is the load of pollutant ( $\text{g y}^{-1}$ ) arising from NAPL source  $k$ , and  $A$  is the area of the city ( $\text{m}^2$ ).

There is a lot of information needed to apply this approach! In principle, information is needed on every source of pollution, i.e. on every parcel of land with potentially polluting activities, every leak in sewers and drains, and every NAPL spill. This detail clearly is not available. An alternative is to group sources, and to consider typical or average conditions for each one. Thus parameter values for a typical petrol station, an average dry cleaner and a representative park could be used, together with counts or estimates of the total number and size of each group.

### Integrated approach

The integrated approach recognises the lack of data available to estimate the individual components. It looks at the final impact on groundwater, and uses a mass balance concept to back-calculate the total load from measured concentrations in groundwater. If pollution has been continuing for long enough, the groundwater system will have reached a steady state. Then incoming pollutant loads will equal the rate of discharge of pollutants and the load can be estimated from:

$$L = \frac{\sum_{\text{Outflows}} Q_l C_l}{A} \quad (2)$$

where  $Q_l$ ,  $C_l$  are the flow rate ( $\text{m}^3 \text{y}^{-1}$ ) and concentration ( $\text{g m}^{-3}$ ) in aquifer outflow  $l$ .

This approach can be applied at various scales. It can be used with a whole aquifer, the catchment of a spring or, more usefully in urban areas, for the catchment of a borehole. This offers the advantages of simplicity and omission of the uncertainty associated with each step of the component calculation. The disadvantages are the indirect method of interpretation which is used and the assumption of steady state.

An estimate of the time required to reach steady state can be obtained by considering the turnover time of a

catchment. The proportion,  $f$ , of a catchment contributing to discharge by time  $t$  is:

$$f = 1 - \exp(-Rt/bn) \quad (3)$$

where  $R$  is recharge rate ( $\text{m y}^{-1}$ ),  $b$ ,  $n$  are saturated thickness (m) and kinematic porosity (fraction) (Lerner 1992). Taking turnover time to be when 80% of the catchment is contributing to the discharge leads to an estimate of turnover time  $T$  (y) for an ideal aquifer, i.e. one with homogeneous and isotropic properties, uniform and steady recharge, and constant discharge as:

$$T = (u\theta + 1.6bn) / R \quad (4)$$

where  $u$  is the thickness of the unsaturated zone (m), and  $\theta$  (fraction) the water content of the unsaturated zone. The first term is for flow through the unsaturated zone from the surface to the water table and the second term is for flow below the water table to the outflow point, from Equation (3) (Lerner, 2003).

In principle, a transient version of Equation (2), as given by Lerner and Papatolios (1993), could be used for non-steady conditions. However, the transient version requires knowledge of the location of pollutant sources relative to the outflow position. The information requirement would be similar to that of the component approach described above, and so an analytical approach to the integrated method would offer little advantage for transient analysis.

## Case studies

### Nottingham hydrogeology

Adaptations of both approaches have been applied in Nottingham, where a range of studies of urban groundwater have been conducted (e.g. Yang *et al.*, 1999; Taylor *et al.*, 2006; Chisala *et al.*, 2007; Trowsdale and Lerner, 2007). These provide an understanding of the hydrogeology and information on landuses and groundwater pollution.

The hydrogeology of the Nottingham aquifer is summarised by Trowsdale and Lerner (2003). Groundwater is principally found in the Triassic Sherwood Sandstone Group. This is a fluvialite red-bed sequence which varies in thickness from zero in the west to over 150 m in the north. Regional hydraulic conductivity is typically a few  $\text{m d}^{-1}$ . Matrix porosity is about 25–35%, specific yield about 10–15%, and there is some fracturing. It has no significant superficial cover to complicate interpretation. 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 19<sup>th</sup> century, and is continued by a network of one urban and six rural pumping stations, supplemented by surface water from the River Derwent.

### The integrated approach: nitrogen loads in Nottingham

The Nottingham aquifer is relatively thick and has high porosity, giving it a long turnover time. Taking

representative values for the parameters in Equation (4) ( $u=25$  m,  $\theta=0.1$ ,  $R=0.2$  m  $y^{-1}$ ,  $n=0.2$ ,  $b=80$  m) gives a turnover time of 140 y. Nitrogen (N) loads have been increasing over this period and the steady state integrated approach cannot be used to estimate N loads from groundwater samples. Instead, a transient analysis was carried out using a groundwater flow model (Lerner *et al.* 1999).

The analysis had two principal steps:

1. A transient groundwater flow model was built and calibrated against groundwater head measurements. This provided an overall water balance and particularly estimates of recharge for each zone. A flow model is prerequisite for a solute transport model, as developed in step 2.
2. A regional solute transport model for N was built and calibrated against observations of concentrations in pumped boreholes. This provided estimates of the concentrations of N in recharge for each zone for all time periods.

The analysis suggested that the average load of N to groundwater over the city of Nottingham is 2.1 g  $m^{-2}$   $y^{-1}$ . This is comparable with loads under rural agricultural areas, and shows that urban areas have significant non-agricultural sources of N loads to groundwater.

An extension of the analysis gave some breakdown of the components of the N load. It used information from similar models of  $SO_4$  and Cl load

#### **Borehole Optimisation System**

A tool called BOS (Borehole Optimisation System) has been developed to assess the risks of pollution to boreholes in urban areas, using Nottingham as a case study (Tait *et al.*, 2004; Chisala *et al.*, 2007; Tait *et al.*, 2008). It contains a groundwater flow model and a comprehensive GIS database of land parcels (1900–1990) on the unconfined aquifer. For each of the ~16 000 parcels of industrial or potentially contaminated land, the database contains the type of industry, the chemicals associated with that industry, and the dates when the site was operational. Most of the sites have not been investigated for pollution. Even if they had, such information is commercially sensitive and would not be readily available. An alternative approach is needed to allocate loads to each parcel.

A standard conceptual model was applied to all parcels to estimate their load of organic industrial pollutants such as PCE (tetrachloroethene, a common chlorinated solvents and degreasing agent) and MTBE (methyl tertiary butyl ether, an octane-enhancing additive to petrol). The pollutant was assumed to be present in a mixed NAPL, and to be dissolving in recharge; the size of the source was taken to be proportional to the size of the site. On this basis, and analogously with the first term in Equation (1), the load from an individual parcel is:

$$L_i = R_i \alpha A_i S \quad (5)$$

where  $S$  is the effective solubility of the pollutant in a typical NAPL,  $R$  is the recharge rate,  $\alpha$  is the proportion of the site that is a pollutant source, and  $A$  is the area of the parcel.

To allow for the lack of information on how severely individual sites are polluting, a stochastic element was included, with  $\pm$  values being drawn randomly from a uniform distribution in the range 0–1. In order to estimate the concentration at a borehole, BOS includes a probabilistic fate and transport model, which takes travel times from the underlying groundwater flow in BOS and incorporates sorption and biodegradation in an analytical solution to calculate concentrations arising from all the possible sources in the borehole's catchment. In addition, validation exercises were undertaken for each study, comparing BOS predictions with the observed concentrations in existing boreholes.

#### **The component approach: industrial pollutants in Nottingham**

One of the applications of BOS was to estimate the risk arising from MTBE (Chisala *et al.*, 2007). There was very little data available on MTBE concentrations in Nottingham groundwater, and so the risk model was validated against a national database of MTBE concentrations. If the model is accepted as accurate, the current MTBE load to unconfined groundwater in Nottingham is 1 g  $m^{-2}$   $y^{-1}$ . It is difficult to know whether this is a reasonable value, but a rough estimate can be made as follows. Petrol contains about ~5% MTBE, so this load implies a loss of ~2000  $m^3$   $y^{-1}$  of petrol over the city, or, assuming 50 petrol stations in Nottingham, about one tanker-full per petrol station per year; this may be a bit high. However, for the fate and transport model in BOS, the biodegradation rate and the load are related, i.e. if the assumed load is increased, the biodegradation rate must also be increased to yield the same predicted concentrations. The biodegradation rates used in BOS were based on a literature review and taken to be uniformly distributed between 0.00035 and 0.0035  $d^{-1}$ , with an average rate of 0.0019  $d^{-1}$ . If the rate was reduced by an order of magnitude, then the estimated load would also be reduced by a factor of 10.

#### **The Birmingham aquifer**

The Birmingham aquifer has been studied as intensively as the Nottingham one (e.g. Ford and Tellam, 1994; Rivett *et al.*, 1992; Rivett *et al.*, 2005). Data from two of the recent studies can be used to derive partial estimates of pollutant loads.

Thomas and Tellam (2006) have initiated a component analysis of recharge and loads, building from a GIS database of landuse on the aquifer. They have developed a model of rainfall recharge for each land parcel which takes account of the effects of urbanisation. They propose that non-point loads of pollution can be estimated by combining these recharge rates with *event mean concentrations* (EMCs). EMCs are the average concentration expected for a particular land use, climate and geology; they give some suggested values taken from unpublished sources and literature reviews.

Combining Thomas and Tellam's (2006) recharge rates with EMCs for nitrate gives a estimated average load to groundwater of 0.3 g  $m^{-2}$   $y^{-1}$  (John Tellam and Abraham Thomas, *pers. comms.*, May 2008). This is only for non-point sources, and excludes leaking sewers, point sources, contaminated land, etc. Nevertheless, it is comparable with the 0.19 g  $m^{-2}$   $y^{-1}$  (9% of 2.1 g  $m^{-2}$   $y^{-1}$ ) for soil leaching

estimated for the average over the whole of Nottingham.

Rivett *et al.* (2005) have provided the information that allows an integrated analysis of chlorinated solvents loads in the catchments of pumped boreholes in the Birmingham aquifer. They have presented solvent concentrations in pumped borehole discharges in both 1987 and 1998, and estimated the proportion of the aquifer's recharge that is being removed by these boreholes (43% in 1987, 5% in 1998). The mass of solvent being removed was 1811 kg y<sup>-1</sup> in 1987 and 228 kg y<sup>-1</sup> in 1998. Assuming steady state conditions, and that the concentrations being pumped are representative of aquifer conditions, then these removal rates are equivalent to loads of 0.04 to 0.1 g m<sup>-2</sup> y<sup>-1</sup>.

## Discussion and conclusions

Urban groundwater has value, especially as pressure on water resources increases. A value that is threatened by pollution. Much work has been done to reduce point source loads and clean up historically polluted sites. However, there is a multiplicity of area, point and linear sources which are not properly characterised and often not known about beyond the possibility that they exist, including leaking sewers, retail premises, industrial workshops and fuel storage tanks. The pollution from these sources is so widespread it can be considered to be diffuse urban pollution.

There are two conceptually different approaches to estimating such loads. The *component* approach considers the load from each land parcel. This approach requires a large amount of data, not only on the location of all land parcels but also on the actual or likely quantity of pollution emanating from them. Such data on pollution are unlikely to be available and the component approach is more likely to be used in a simplified form, considering the loads from each category of land parcels. This allows the use of good quality location data, which is fairly easy to obtain from historical maps and records, together with generic information on pollutants loads associated with each category of source.

The alternative approach to estimating loads is the *integrated* approach. This takes the observed concentrations in groundwater discharges to represent an integration of the loads on the catchment area feeding that discharge. It requires much less information than the component approach, but has greater assumptions, for example that steady state has been reached. This assumption can be tested by assessing the turnover time for the catchment of the discharge point being examined.

Nitrogen loads have been estimated for the Nottingham and Birmingham aquifers in the UK. The integrated approach was used in Nottingham, and provided an estimate of the total N load as 2.1 g m<sup>-2</sup> y<sup>-1</sup>, of which 0.19 g m<sup>-2</sup> y<sup>-1</sup> was from non-point sources. A component analysis for Birmingham gave a comparable non-point N load of 0.3 g m<sup>-2</sup> y<sup>-1</sup>; all loads are averaged over the whole aquifer.

Loads of selected industrial pollutants were also estimated for Nottingham and Birmingham. A component analysis for MTBE in Nottingham estimates a load of 1 g m<sup>-2</sup> y<sup>-1</sup>. There is substantial uncertainty in this load estimate as it is dependant on the assumed biodegradation rate, which is also unknown. In Birmingham, an integrated analysis, using concentrations in pumped boreholes,

estimated that the load of chlorinated solvents was in the range 0.04 to 0.1 g m<sup>-2</sup> y<sup>-1</sup>. Chlorinated solvents are mainly spilt as DNAPLs, and the load estimate is not the amount of DNAPL spilt but the transfer (dissolution) rate from DNAPL to groundwater. Comparison of the loads for the two types of pollutant suggests that the MTBE estimate may be too high, particularly considering the uncertainties caused by the poor information on biodegradation rates and the absence of any direct measurements of concentrations.

Very little attention has been paid to the loads and impacts of such diffuse pollution. I have been unable to find any other information which can be used to estimate urban pollutant loads on groundwater, despite the importance of urban groundwater for water supply and environmental flows. Sustainable urban hydrology must consider how to estimate such loads, and how to design urban infrastructures better to reduce the loads to acceptable levels, whatever they may be. There is plenty of scope for further work in this area!

## References

- Chisala, B.N., Tait, N.G. and Lerner, D.N., 2007. Evaluating the risk of methyl tertiary- butyl ether (MTBE) to urban groundwater at city scale: Nottingham case study. *J. Contam. Hydrol.*, **91**, 128–145.
- Ford, M. and Tellam, J.H., (1994). Source, type of extent of inorganic contamination within the Birmingham urban aquifer system, UK. *J. Hydrol.*, **156**, 101–35.
- Lerner, D.N., 1992. Borehole catchments and time-of-travel zones in aquifers with recharge. *Water Resour. Res.*, **28**, 2621–2628.
- Lerner, D.N., 2003. Estimating urban loads of nitrogen to groundwater. *J. CIWEM*. **17**, 239–244.
- Lerner, D.N. and Papatolios, K.T., 1993. A simple analytical approach for predicting nitrate concentrations in pumped ground water. *Ground Water*, **31**, 370–375.
- Lerner, D.N., Yang, Y., Barrett, M.H. and Tellam, J.H., 1999. Loadings of non-agricultural nitrogen in urban groundwater. Impacts of urban growth on surface and ground waters (Proc. IAHS symposium HS5, Birmingham, July 1999). *IAHS publication no.259*, 117–123.
- Rivett, M.O., Lerner, D.N., Lloyd, J.W. and Clark, L., 1990. Organic contamination of the Birmingham aquifer. *J. Hydrol.*, **113**, 307–23.
- Rivett, M.O., Shepherd, K.A., Keeys, L. and Brennan, A.E., 2005. Chlorinated solvents in the Birmingham aquifer, UK: 1986–2001. *Quart. J. Eng. Geol. Hydrogeol.*, **38**, 337–50.
- Tait, N.G., Davison, R.M., Whittaker, J. J., Leharne, S.A. and Lerner, D.N., 2004. Borehole Optimisation System (BOS) - A GIS based risk analysis tool for optimising the use of urban groundwater. *Environ. Model. Softw.*, **19**, 1111–1124.
- Tait, N.G., Davison, R.M., Leharne, S.A. and Lerner, D.N., 2008. Borehole Optimisation System (BOS) - A case study assessing options for abstraction of urban groundwater in Nottingham, UK. *Environ. Model. Softw.*, **23**, 611–621.

- Taylor, R.G., Cronin, A.A., Lerner, D.N., Tellam, J.H., Bottrell, S.H., Rueedi, J. and Barrett, M.H., 2006. Hydrochemical evidence of the depth of penetration of anthropogenic recharge in sandstone aquifers underlying two mature cities in the UK. *Appl. Geochem.*, **21**, 1570–1592.
- Thomas, A. and Tellam, J., 2006. Modelling of recharge and pollutant fluxes to urban groundwaters. *Sci. Tot. Environ.*, **360**, 158–179.
- Trowsdale, S.A. and Lerner, D.N., 2003. Implications of flow patterns in the sandstone aquifer beneath the mature conurbation of Nottingham for source protection. *Quart. J. Eng. Geol. Hydrogeol.*, **36**, 197–206.
- Trowsdale, S.A. and Lerner, D.N., 2007. Assessing the origin and age of urban ground water with depth - a modelling approach. *J. Contam. Hydrol.*, **91**, 171–183.
- Yang, Y., Lerner, D.N., Barrett, M.H. and Tellam, J.H., 1999. Quantification of groundwater recharge in the city of Nottingham, UK. *Environ. Geol.*, **38**, 183–198.

