Simazine concentrations in soil, groundwater and stream water following application to *Pinus* plantations in the coastal lowlands of south-east Queensland.

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Summary

A study was conducted to assess the fate of simazine applied to exotic *Pinus* plantations in the coastal lowlands of south-east Queensland. The study had two components, the first being a large-scale catchment study site (903 ha) which assessed the level of simazine residues being transported from the point of application to the shallow unconfined aquifer and the major drainage stream over a 13-month period. The second component consisted of a separate study on a small-scale study site (0.1 ha) to assess simazine persistence and its potential to leach in the coarse textured and relatively infertile soils of the area.

An 80 ha clearfelled area within the large-scale study catchment was re-established with a second rotation plantation and routinely treated with simazine at 2.5 kg a.i. ha^{-1} over an 11-month period. A stream monitoring station at the base of the catchment sampled streamwater, and two piezometers provided access for groundwater monitoring. At the small-scale study site, simazine was applied to small plots at the above rate and the soil profile and shallow groundwater were routinely sampled.

Simazine was regularly detected in streamwater after surface runoff events but was below the current Australian drinking water health value. The detection of simazine in the unconfined aquifers at both sites indicated that it has the potential to leach to groundwater. However, under routine applications it would seem that the groundwater concentrations were low and short-lived (persistence < 6 weeks). The results indicated that simazine has a relatively short half-life (mean 13 days) in the coarse textured soils of the coastal lowlands of south-east Queensland. Transport from point of application by runoff or leaching was thought to be a minor pathway for loss from these plantations. Recommendations for future monitoring and research directions are discussed.

Introduction

Simazine (2-chloro-4,6-bis-[‘ethylamino]-s-triazine) is a triazine compound which is used in Queensland Department of Primary Industries Forestry (QDPIF) managed *Pinus* plantations as a residual herbicide. In many countries including the USA, simazine and other triazine compounds (e.g. atrazine) have been reported to be major groundwater contaminants and thus have received a great deal of attention from regulatory bodies (Zhang *et al*. 1997). Consequently, triazine compounds have been widely monitored in horticulture and broad acre agriculture and forest plantations because of the potential for residues to be detected in surface water and groundwater reserves (Hassall 1990).

Simazine has a relatively low water solubility (5 mg L^{-1} at 20 °C) with a modest affinity to sorb to soil organic matter as demonstrated through an organic carbon to water partition coefficient (Koc) of 135 g mL^{-1}. Mobility in soil ranges from slight in clays to high in sandy loams (EPA 1998). Microbial decomposition and hydrolysis are generally recognised as the major degradation pathways in soil. Photolysis can also be an important loss mechanism at or above the soil surface, however following simazine incorporation (i.e. through rainfall, irrigation, and tillage) into the soil profile loss through this mechanism is practicably nil, as is the loss through volatilisation (WSSA 1994). Runoff and leaching are considered to be an important means of transport from point of application (Yaron *et al*. 1985).

The QDPIF *Pinus* plantation estate (130 000 ha) is primarily located in the coastal lowland areas of Queensland with the majority (100 000 ha) located in the south-east of the State. In general, the topography of these areas is flat to undulating (slopes < 5%) with an elevation above sea level (a.s.l.) around 50 m. Typically, soils are coarse textured, infertile with low soil organic carbon and often overlie a clay aquitard which is found at depths between 0.5 and 10 m. Prolonged waterlogging of the upper soil profile can occur in areas where the aquitard is within 2 to 3 m of the surface, particularly during years of average and above rainfall when a seasonal perched groundwater table often exists for periods up to 6 months (Bubb and Croton, 2001). In areas prone to waterlogging, site preparation consists of either continuous mounding or spot mounding, whereas in drier areas either strip cultivation or wing ripping is carried out.

The use of simazine as a residual herbicide is an important cost effective means of controlling fast growing competitor weed species in sub-tropical and tropical Queensland *Pinus* plantations. During the first year of plantation establishment, simazine is usually applied three times (dependent on weed development) at a rate of 2.5 kg a.i. ha^{-1} either by tractor or manually. Application of herbicide is excluded from major drainage features within plantations in line with current watercourse protection guidelines (see Costantini *et al*. 1993). These require a retained riparian zone (between 10 and 30 m depending on watercourse classification) around ephemeral and permanent streams, as well as maintaining stable waterways to receive runoff from mound furrows and roading.

To date most research investigating the environmental fate of simazine has been carried out overseas, with the limited published research in Australia being mostly directed to atrazine. Because simazine is expected to be reasonably mobile in the coarse textured and relatively low organic carbon soils common to the QDPIF Pinus estate in the coastal lowlands, there is a risk of its movement into groundwater and stream water following application to these forest plantations. This study investigates the effectiveness of current QDPIF forest management practices in protecting catchment water quality values, and provides feedback for current best management practices.

Methods

Study Sites

The study was conducted within State owned and QDPIF managed *Pinus* plantations at Toolara (25° 55' S, 152° 50' E) situated approximately 200 km north of Brisbane, Queensland. The area has a sub-tropical climate with an annual average rainfall and pan evaporation of 1312 and 1430 mm respectively, with mean temperatures between 12.7 (winter) and 25.7 °C (summer). Typically rainfall is summer dominant with the wettest period of the year being usually between January and March.

The study had two components, the first a large-scale study which was conducted within an existing long-term catchment study, and the second a separate small-scale study conducted on small-plots in an adjoining catchment. The topography surrounding the study sites is flat to gently inclined and around 75 m a.s.l.. The underlying geology consists of freshwater sediments accumulated during the early Mesozoic and an overlying mantle of Quaternary siliceous sands (Coaldrake 1961).

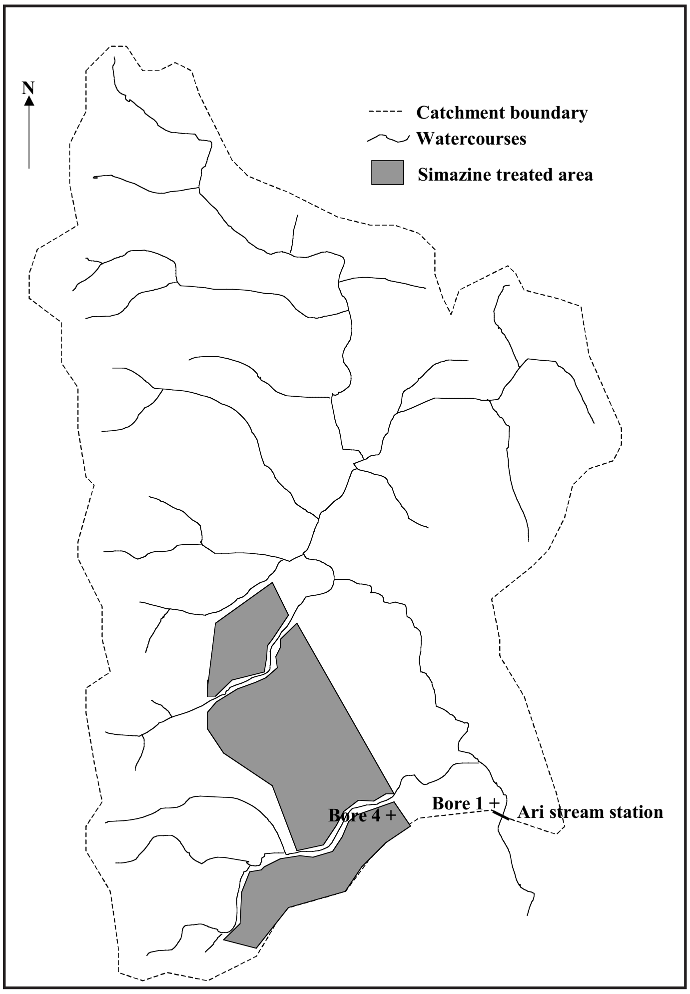
The main soil types within the large-scale study catchment are Siliceous Sands, Lithosols, Grey and Gleyed Podzolics, Yellow Earths, Yellow Podzolics, and Humus Podzols (after Stace *et al.* 1968). With the exception of areas adjacent to drainage channels, the soils are generally deep with no obvious aquitard between the surface and the sandstone basement rock at a depth around 10 to 15 m. Exploratory bores in the large-scale study suggest an unconfined aquifer is contained within the soil profile above the sandstone, and piezometric levels suggest that groundwater is generally present all year round at depths between 2 and 12 m. In contrast, the soil at the small-scale leaching site is shallow, being a Gleyed Podzolic above a heavy clay aquitard at a depth around 1.5 m. A shallow perched watertable forms above this aquitard for several months of the year. The surface soils are subjected to considerably longer periods of waterlogging than those at the large-scale study site where waterlogging is restricted to areas around the main drainage features. Research by Bubb and Croton (2001) reported that the primary process for the removal of groundwater in the study area was considered to be via evapotranspiration, as little lateral or vertical drainage occurred because of the low slope and low saturated hydraulic conductivity of the lower soil profile. Typically, streams are emphemeral, being characterised for much of the year by a series of permanent waterholes distributed along their length. During average rainfall years, streamflow usually occurs for the several months coinciding with peak rainfall, whereas in below average rainfall years, zero streamflow is not uncommon.

Treatments and Monitoring

Large-scale study

The large-scale study was conducted in the upper reaches of Sandy Creek within a 903 ha catchment containing 810 ha of *Pinus elliottii* and *P caribaea var. hondurensis* plantations. At the base of the catchment a triangular profile flat-V weir (hereafter referred to as Ari weir) was constructed and the stream station instrumented with data loggers, automatic sampler, and an *in situ* streamheight sensor (Figure 1). A stage-discharge relationship was derived from Bos (1989) and periodically revised through manual flow gauging; this included large flood events when non-modular flow was evident at the weir. Groundwater was sampled from two exiting piezometers located in the study area (installed 1996), one located within the treated area to monitor vertical movement of simazine (Bore 4), and the other adjacent to the stream station to monitor the lateral movement of simazine from the point of application (Bore 1). Bores were hollow augured to the sandstone basement rock, following installation of the casing were backfilled with coarse sand along the screened section (lower 3 to 6 m), followed by bentonite with an upper collar of cement to the ground surface where the exposed casing (0.4 m) was capped.

Figure 1. Layout of the large-scale study within a 903 ha catchment in the Toolara *Pinus* plantation estate.



A harvest treatment (80 ha clearfall and 250 ha thinning) was implemented in 1998. The clearfall area was subsequently prepared for planting and the second rotation established later that year. The proportion of the catchment re-established with second rotation (9 %) was considerably greater than the broad average for catchments containing the plantation estate (< 3 %) based on a 30-year rotation and taking into account catchment areas not under plantation such as, roading, stable waterways and retained riparian zones. Simazine was routinely applied to the second rotation area on six occasions from November 1998 to September 1999 (Table 1) at a rate of 5 kg a.i. ha^{-1} (treated), the treated area being a 2.5 m band between 5 m rows which was effectively 50 % of the plantation area (i.e. the actual broad-scale rate was 2.5 kg a.i. ha^{-1}). During the first two applications in November 1998 and January 1999 the entire treated area was tended in a single operation, whereas during the period April to September 1999 the treated area was subdivided and progressively tended in four separate operations.

Table 1. Schedule of simazine applications to the large-scale study site

Application Period	10/11/98 to 16/11/98	6/1/99 to 12/1/99	19/4/99 to 30/4/99	7/7/99 to 23/7/99	13/8/99 to 20/8/99	10/9/99 to 14/9/99
[1]Treated area (ha)	40	40	12	9	12	7

[1] Herbicide is applied in a band to 50 % of the plantation, hence 'treated area' represents half of the established plantation area.

Small-scale study

The small-scale leaching study was based on a design incorporating four replications of a single simazine rate. Plot sizes were 50 x 3 m with a minimum buffer width of 4 m between plots. No surface debris from the previous rotation or harvest operations was retained on this site. Simazine was applied on 6th August 1998, 11th November 1998 and 25th March 1999 (hereafter referred to as application 1, 2 and 3 respectively) at a rate of 5 kg a.i. ha^{-1} in liquid spray formulation, by knapsack as either a fan or sprinkler spray, along a band width of 3 m. All spray applications were performed using QDPIF current prescribed best practice. Immediately prior to application 3, a piezometer was installed in each plot to allow sampling of groundwater from the shallow perched aquifer system. Piezometers were installed to immediately above the clay aquitard (screened along entire submerged casing except for the upper 0.3 m) in a similar manner as outlined above. The bores were purged numerous times during construction as groundwater levels were within 0.5 m of the surface.

Sampling

Large-scale study

A combination of grab and automated event samples were collected from the headwaters of Ari weir. Grab samples were generally taken at fortnightly intervals, whilst automatic water samples were taken at designated stream heights throughout a flood event. High purity solvent-rinsed glass bottles were used to collect samples, with automated water samples being drawn from a teflon coated stream intake line which was purged prior to sampling. Following removal of samples from the automated sampler, distilled water was purged through the intake line and sampler to provide a sample blank for any residual contamination within the system. A grab sample was also taken from the weir headwaters immediately prior to simazine application. Samples were stored at 5°C in a chilled portable cool chest and transported to the analytical laboratory.

Groundwater sampling was undertaken on a monthly basis using a small electrical pump with a teflon delivery line. Piezometers were purged prior to sampling, samples were then collected, stored and transported in a similar manner as outlined above.

Small-scale study

Following individual simazine applications, soil samples were collected at regular intervals over a 3-month period to assess the leaching and degradation pattern of simazine. In general, sampling was scheduled for 1, 7, 14, 28, 56 and 112 days after treatment.

Soil samples were taken from the following depth intervals: 0-10; 10-20; 20-30; 30-45; 45-60; 60-90 cm. From each interval, four samples were taken and subsequently bulked and then subsampled to provide a 500 g sample. Soil samples were wrapped in alfoil and transferred to portable cool chests for transporting to a freezer storage prior to analysis. Soil bulk density was determined from the mean of five replicate samples taken at each depth interval from a soil pit adjoining the study plots using a standard core method. Sampling of groundwater from the piezometers was carried out generally after a rainfall event which exceeded 25 mm. The methods used were similar to those outlined above for the groundwater sampling in the large-scale study.

Laboratory methods

Samples were analysed for simazine by high performance liquid chromatography. Water samples were assayed using the method of Reupert *et al.* (1990) and soil samples by the method of Walker and Blacklow (1994). Simazine soil concentration was transformed to units of kg ha^{-1} using bulk density measurements outlined above.

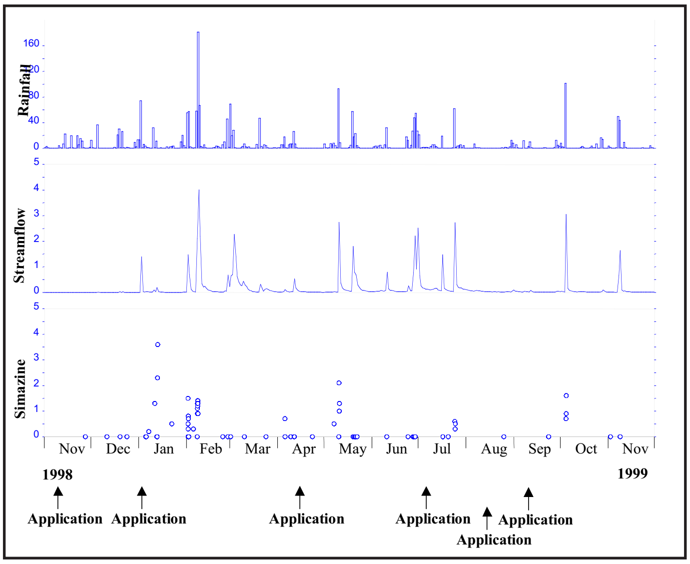
Results

Large-scale study

The total rainfall (2203 mm) during the 13-month study reflected a period of higher than average rainfall, the long-term annual mean being 1312 mm. This and the harvest treatment were responsible for near continuous streamflow during the study period, in particular after early January 1999. This pattern of streamflow is in contrast to the previous 5 years where flow was limited to less than 2 months annually (data not presented).

Analysis of grab samples indicated that during periods of baseflow or zero flow, streamwater concentration of simazine was below limits of detection (with exception of the sample taken on 22/1/99). In contrast, residues were detected in streamwater during seven flood events over the study period, including the major flood event that occurred in February 1999 (Figure 2). In general, after each individual application, simazine could be detected in subsequent flood events for up to six weeks. Streamwater concentration did not exceed the NHMRC health value (20 $\mu g\ L^{-1}$), however the guideline value (0.5 $\mu g\ L^{-1}$) was exceeded for a total of 235 hr during the seven flood events which represents 20.4 % of streamflow during the study period. Regression analyses revealed a poor relationship (R^2 = 0.02, $p < 0.05$) between the concentration of suspended solids and simazine during flood events.

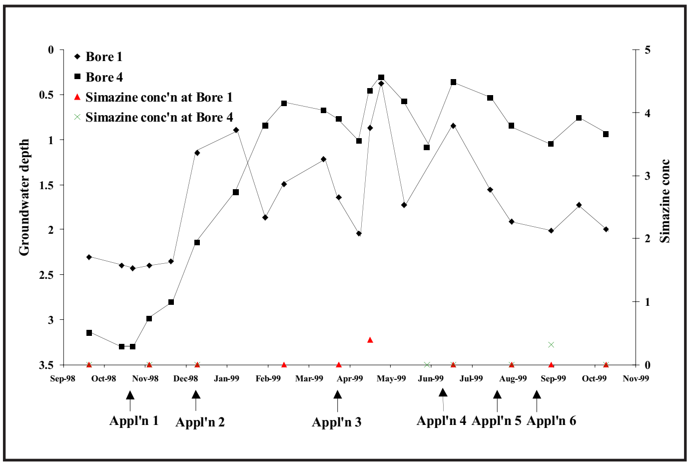
Figure 2. Rainfall (mm d^{-1}), daily streamflow (cumecs) and simazine concentration (μg L^{-1}) at catchment outlet.



An estimate of the simazine flux in streamflow was calculated to gain an understanding of the potential mass being transported from the point of application to the main drainage stream at the large-scale site. The flux was estimated by summing the product of simazine concentration and discharge during the time period associated with the sample, the time period being the mid-point from the previous sample to the mid-point of the following sample. This yielded a flux of 0.6 kg which represented 0.5 % of the mass applied to the plantation (i.e. 120 kg).

Simazine was not detected in groundwater at either bore during the study period with the exception of samples taken on 13/5/99 (Bore 1) and 23/9/99 (Bore 4) (Figure 3). Furthermore, the 'positive' samples were low, being between 0.3 and 0.4 μg L^{-1} respectively. The near surface depth of groundwater in both wells was typical of catchment response to clearfelling in the coastal lowlands of south-east Queensland.

Figure 3. Groundwater depth (m) and simazine concentration (μg L^{-1}) in Ari catchment.



Small-scale study

Results from the three separate applications indicated that the residual effect of simazine in the soil profile was short (Tables 2 to 4). Based on first order kinetics the calculated half life for applications 1, 2, and 3 was 25, 5 and 10 days respectively (mean 13 days). Following application 1, simazine was detected in the soil to a depth of 45 cm after 15 days, whereas following applications 2 and 3, detectable levels were confined to the upper 20 cm. Groundwater samples taken from the shallow perched aquifer after application 3 contained low concentrations of simazine (i.e. < 0.7 mg L-1) for a period less than 43 days after treatment (Table 5).

Table 2. Mean simazine residues (kg ha^{-1}) in soil following application 1 (6th August 1998).

DAT[a] / Depth (cm)	1	8	15	39	57	96
0-10	4.15	2.98	2.83	2.50	0.73	0.50
10-20	1.10	1.45	0.70	0.55	0.30	n.d.
20-30	0.60	0.83	0.50	n.d.	n.d.	n.d.
30-45	n.d.	0.60	0.70	n.d.	n.d.	n.d.
45-60	n.s.	n.s.	n.s.	n.d.	n.d.	n.d.
60-90	n.s.	n.s.	n.s.	n.d.	n.d.	n.d.
∑ Rainfall since application (mm)	9.2	9.2	15.2	150.0	180.8	241.8

[a]Days after treatment; n.d. not detected; n.s. not sampled

Table 3. Mean simazine residues (kg ha^{-1}) in soil following application 2 (11th November 1998).

DAT[a] / Depth (cm)	7	14	28	56
0-10	2.40	0.70	n.d.	n.d.
10-20	0.60	n.d.	n.d.	n.d.
20-30	n.d.	n.d.	n.d.	n.d.
30-45	n.d.	n.d.	n.d.	n.d.
45-60	n.s.	n.d.	n.d.	n.d.
60-90	n.s.	n.s.	n.d.	n.d.
∑ Rainfall since application (mm)	32.2	134.0	180.6	389.6

[a]Days after treatment; n.d. not detected; n.s. not sampled

Table 4. Mean simazine residues (kg ha^{-1}) in soil following application 3 (25th March 1999).

DAT[a] / Depth (cm)	1	14	28
0-10	5.40	3.30	1.10
10-20	0.70	0.50	n.d.
20-30	n.d.	n.d.	n.d.
30-45	n.d.	n.d.	n.d.
45-60	n.s.	n.d.	n.d.
60-90	n.s.	n.s.	n.d.
∑ Rainfall since application (mm)	1.2	15.4	55.6

[a]Days after treatment; n.d. not detected; n.s. not sampled

Table 5. Mean simazine residues in perched aquifer at the small-scale study site following application 3 (25th march 1999).

DAT[a]	Σ rainfall (mm)	Groundwater depth (m)	Simazine (μg L^{-1})
-1	0	0.31	<0.2
14	33	0.70	0.7
21	52	0.43	0.6
43	27	0.69	<0.2
53	108	0.36	<0.2

[a]Days after treatment

Discussion

The detection of simazine during flood events, rather than through periods of baseflow or zero flow, indicates that transport to streamwater was primarily associated with surface runoff. Presumably transport in runoff was associated with the water phase rather than the solid phase as suggested by the rainfall simulation study on sandy loam soils reported by Gouy *et al*. (1999). This presumption is also supported indirectly by Bubb (1999) who found the movement of the 'sister' compound atrazine in fine textured soils of QDPIF *Araucaria* plantations was associated with the water phase. The poor relationship between suspended solids and simazine concentration during flood events also suggests that simazine movement in the water phase was a more important transport mechanism than via entrained sediment. However, some degree of caution must be taken with this particular observation as the major source of suspended solids in the stream was likely to be derived from roads and associated drains and therefore a high proportion of the entrained sediments delivered to the stream may not have been exposed to simazine application. Nevertheless, because simazine is associated with the water phase, erosion mitigation systems used in QDPIF *Pinus* plantations such as stable waterways and retained riparian zones which augment the deposition of entrained sediment, may be somewhat limited in their ability to minimise solute transport. Furthermore, the waterlogged conditions which commonly prevail in these mitigation systems result in reduced infiltration capacities (Loch *et al*. 1999) and therefore may limit on-site containment of runoff.

The relatively low simazine flux in streamwater (0.5 % of total applied) as compared with the total mass applied to the site indicated that surface runoff was a minor loss pathway from these plantations; this is consistent with other studies (Yaron *et al*. 1985). The estimated stream flux also highlighted the low mass of simazine required for residues to be detected in surface water.

The results indicated that simazine has a relatively short half-life (mean 13 days) in the *Pinus* plantations soils in the coastal lowlands of south-east Queensland. Indeed this finding has prompted research into improving the efficacy of simazine through refined application techniques, as well as reviewing alternative herbicides. In comparison, reported half life from overseas studies range between 28 and 149 days (Perez *et al*. 1997; Wauchope *et al*. 1992). The half life for simazine is similar to that reported by Bubb (1999) for atrazine in both QDPIF *Araucaria* and *Pinus* plantations in south-east Queensland. The major disparity in soil characteristics between these plantations (Araucaria plantation soils are typically fine textured with high soil organic carbon), yet close proximity and similar climatic patterns, suggests half life is independent of soil physicochemical conditions.

The detection of simazine in the lower soil profile and the shallow aquifer system indicated its potential to leach in the coarse textured and low soil organic carbon soils of Queensland's coastal lowlands. This is in line with findings by Kruger et al. (1996) who found the mobility of simazine in soils to be negatively correlated with soil organic carbon and positively correlated with sand content. Nevertheless, under the simazine prescriptions routinely used by QDPIF in the study area, it would seem that groundwater concentrations in the shallow aquifers are low and short-lived (persistence < 6 weeks). The absence of a significant mass of simazine in the lower soil profile and groundwater suggests that leaching is not a major loss pathway. Further to this, the peak mass of simazine contained within the perched aquifer at the small-scale site occurred at 21 days after treatment. An estimate of this mass contained in the aquifer based on the groundwater concentration and groundwater volume (depth x specific yield) accounted for only 1 g ha^{-1}. Whether routine application of simazine to these forest plantations would pose a risk to deeper aquifers is largely unknown, particularly as the nature and extent of these aquifers and associated groundwater fluxes are poorly understood. Nevertheless, it is suggested that the likelihood of simazine accumulating in the deeper aquifers is small given the following factors: the low saturated hydraulic conductivity (i.e. 2.0 mm d^{-1}) and thickness (between 2 and 10 m) of the clay aquitard (see Bubb and Croton, 2001); the relatively low loading to the shallow aquifer as demonstrated with this study and; limited loading to any site as simazine is applied only in the first 12 months of a 30 year rotation.

Assuming losses due to volatilisation are minimal as the literature suggests, and that losses by either leaching or surface runoff are similarly low as this study suggests, the major loss pathway for simazine from the study area is likely to be *in situ* degradation (i.e. microbial, hydrolysis). Although photolysis may also be important, the rainfall which occurred at the small-scale study site immediately following each application would have likely incorporated most of the applied simazine into the soil. Thus only a limited amount would have been affected by this loss mechanism.

The simazine residues detected in both surface and groundwater during the 13-month study period were well below the NHMRC health value of 20 μg L^{-1} (NHMRC 1996). Given the above average rainfall and large number of runoff events which occurred during this study there is a low probability of simazine residues exceeding the NHMRC health value on freely draining plantation sites provided current QDPIF management practices are observed. In particular, these practices include the retained riparian zone around watercourses, and redirection of channeled runoff to the general plantation area and stable waterways to maximise on-site containment of runoff. However, it should be stressed that the surface water results reflect sites that have relatively free drainage (i.e. restriction layer > 5 m) and which make up a large proportion of the estate. In contrast, it is expected that sites subjected to prolonged periods of waterlogging (i.e. 30 % of estate) may represent a greater risk to surface water quality because of the higher potential for runoff and hence off-site transport of simazine. Therefore it is important that a simazine monitoring program be directed towards sites which contain a high proportion of periodically waterlogged areas. Additional research should be aimed at investigating: (1) site management practices which contain on-site runoff, for instance, those based on non-furrow mounding techniques (e.g. excavator-based spot mounding) which may promote greater site infiltration capacities over conventional continuous mounding systems; (2) critical width for retained riparian zones in relation to drainage features; (3) improved application techniques which lead to reduced rates of application; (4) alternate cost-effective herbicides. Research is also required at a regional catchment level in regard to the spatial distribution of simazine treated plantations, downstream dilution dynamics and off-site simazine residues.

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