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Environmental fate of terbuthylazine and hexazinone in a planted forest steepland Recent Soil, New Zealand

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Abstract

Background: The use of herbicides for weed control in New Zealand planted forestry is important for primary production. The objective of this study was to assess the environmental fate of terbuthylazine and hexazinone in a Recent Soil following an aerial post-plant herbicide application in a New Zealand planted forest.

Methods: Soil and forest litter were collected from four plots within a recently harvested and replanted *Pinus radiata* experimental sub-catchment. Samples were taken before and up to 6 months after the aerial application of terbuthylazine and hexazinone.

Findings : There was no drainage below 1-m depth in the first month after spray application. On the following month, there were six drainage events ranging from 12 to 36 mm. Both terbuthylazine and hexazinone were located predominantly in the 0–10 cm soil depth, followed by the litter layer with only small amounts detected below 10 cm soil depth. Following a month of drainage events, hexazinone showed an increase at a soil depth of 10–50 cm. Both terbuthylazine and hexazinone rapidly dissipated with time after spray application, and predicted half-lives for both active ingredients were less than 1 day.

Conclusions: The first 2 weeks after spray application were found to pose the greatest potential risk of movement off-site, after which the risks were low. Organic matter management practices that influence the amount and distribution of forest floor, litter and harvest residue have an important role in determining the environmental fate of both terbuthylazine and hexazinone.

Keywords: Planted forest, Herbicide, Soil, Terbuthylazine, Hexazinone, New Zealand

Background

Herbicides are used during establishment of planted forests to effectively control competing vegetation both in New Zealand (Rolando et al. 2013) and internationally (George and Brennan 2002; Wagner et al. 2004; Weatherford et al. 2015). The use of herbicides during the early stages of the rotation significantly enhances forest productivity (Wagner et al. 2006) and is cost effective (George and Brennan 2002; Rolando et al. 2011), making their use important for forest management. There is, however, a growing awareness of the potential environmental impact of herbicides when they are applied to the environment. The global trend within forests to reduce herbicide use (Little et al. 2006; McCarthy et al. 2011; Thompson and Pitt 2003) is driven by national and regional environmental policies and voluntary subscriptions to global sustainable forest management certification schemes, such as the Forest Stewardship Council (FSC) (Rolando et al. 2013). In New Zealand, the majority of forests (56 %) are FSC certified (Forest Owners Association 2014), which requires an overall move away from herbicide use in forests (FSC 2013).

A key to environmental concerns surrounding the use of herbicides is the potential impact on fresh water quality (Baillie et al. 2015; Baillie and Neary 2015). Once applied to the establishing forest, the movement of herbicides into water bodies can occur via surface run-off and leaching or via sediment generation from erosion (Holvoet et al. 2007; Sarmah et al. 2004). The risk period

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for a herbicide moving off-site is determined by the persistence of the herbicide on-site (Garrett et al. 2015).

Terbuthylazine and hexazinone are the two most commonly used active ingredients for post-plant control of forestry weeds within New Zealand's planted forests as they have been found to be most effective amongst the available active ingredients (Rolando et al. 2013). These two chemicals are important tools for vegetation management because they have some residual herbicidal activity unlike some other herbicides (e.g. glyphosate). This property is important for the control of weeds that may germinate from the seed bank. Both of these active ingredients were previously on the FSC list of highly hazardous pesticides between 2007 and 2015 and were unable to be used in FSC-certified forests without a derogation (FSC 2014). These active ingredients were on the FSC list due to the potential for terbuthylazine to bio-accumulate, and for the water solubility and persistence of hexazinone. These active ingredients are now available for use in FSC-certified forests due to a change in the criteria for rating herbicides as highly hazardous. However, there is still uncertainty surrounding the environmental fate of these active ingredients in the New Zealand planted forest environment particularly on soils that are vulnerable to leaching. To maintain best management practices, there is a need to understand the potential for active ingredients to move off-site into water bodies in the New Zealand planted forest environment. The movement down the soil profile and persistence on-site of these active ingredients has been tested in the field on a Pumice Soil (Garrett et al. 2015), which constitutes 25 % of New Zealand's planted forest soils and is considered vulnerable to herbicide movement due to the low organic carbon concentration of the soil (Watt et al. 2010). However, field studies examining movement of these active ingredients in New Zealand planted forests have not been undertaken for other soil types.

The objective of this study was to assess the fate of terbuthylazine and hexazinone in a Recent Soil following an aerial post-plant weed control application of these two herbicides in a New Zealand planted forest catchment. The recent soil was selected as this soil type underlies a significant proportion (13 %) of the New Zealand planted forest estate and is considered vulnerable to herbicide movement due to the low carbon concentration of the soil (Watt et al. 2010).

Methods

Study site and spray application

During 2014, an experimental sub-catchment was selected in Omaio forest (latitude 37° 49′ S, longitude 177° 40′ E) located in the Eastern Bay of Plenty, New Zealand. The sub-catchment was 11.91 ha in size with steep topography (average slope 32°) ranging in elevation from 250 to 400 m a.s.l. The total annual rainfall averaged 1504 mm, and the mean annual temperature was 12.9 °C (NIWA 2012). The mean and maximum spring air temperatures were 12.1 and 16.2 °C, respectively (NIWA 2012). The soil is classified as Typic Tephric Recent Soil in the New Zealand Soil Classification System (Hewitt 1998). The soil parent material originates from fine volcanic ash fall (ranging in depth but approximately 0.5 m thick on average) over sedimentary mudstone. The soil ranges from well to imperfectly drained with sandy loam to sandy clay loam textures in the upper profile and silty clay textures in the lower profile.

The study area was planted with second-rotation *Pinus radiata* D.Don in July 2014 at 750–833 stems per hectare. The previous *P. radiata* crop was harvested in June 2013, and no site preparation was undertaken post-harvest leaving an un-even distribution of forest floor and harvest residues on-site. There were a few soil deposits within the study area caused by small landslides, which had occurred previously as a result of post-harvest erosion events. During April 2014, an operational pre-plant aerial spray (Rolando et al. 2013) was used to control weeds that had regenerated following harvest. This comprised 7 L ha^{-1} of glyphosate (Deal™ 510 RF; 510 g glyphosate L^{-1}; Orion Crop Protection, Auckland), diluted in water to an applied rate of 100 L ha^{-1}. The operational post-planting spray (Rolando et al. 2013) consisted of one aerial application on 10 November 2014 using 15 L ha^{-1} of Agpro Valzine Extra (AGPRO NZ Limited, Auckland, New Zealand; 400 g L^{-1} terbuthylazine and 100 g L^{-1} hexazinone), diluted in water to an applied rate of 100 L ha^{-1}. Tartrazine (Bayer NZ, Ltd) was added to the spray mix (rate 0.0042 kg L^{-1}) as a colorimetric tracer to determine the amount of spray applied (Richardson and Thistle 2006). Very few weeds were present at the time of spray application. Stream water was sampled for the sub-catchment and reported in Baillie 2016.

Four soil plots, with dimensions of 80 m × 20 m, were installed within the experimental sub-catchment avoiding the few small landslides present, and average plot slope was measured. The amount of spray applied to each plot was determined using 60 horizontal sample-collection plates (115.5 cm^2 per plate) positioned along transects within each soil plot to detect the colorimetric tracer. The amount of terbuthylazine and hexazinone active ingredient applied per plot was then calculated based on the density of Agpro Valzine Extra spray mix (AGPRO NZ Limited, Auckland, New Zealand; 1.12 g mL^{-1}) as described in Garrett et al. (2015). The tartrazine-detection results showed that only half of one of the four plots was sprayed; therefore, this half of the plot was excluded from spray calculations and post-spray forest floor and soil sampling.

Collection and analysis of forest floor and soil samples

The methods used for collection and analysis of forest floor and soil samples have been previously described in detail in Garrett et al. (2015). The quantity of coarse woody debris (>10 cm diameter) was measured using the line-transect method of Van Wagner (1968) for volume and mass from oven dry (70 °C) wood density discs for each plot. The percent ground cover by the forest floor (coarse woody debris and litter (<10 cm diameter)) was measured using a transect grid of 400 points over three of the four plots. Litter (LFH; a combination of fresh litter (L), and partly and well-decomposed litter (FH) all <10 cm diameter) and soil chemistry samples were collected before spray application and then at 15, 30, 79 and 163 days after spray application. The litter was collected at four random points per plot using a 0.1-m^2 sampling square and bulked per plot to form one composite sample. Soil samples were collected from 30 random points for 0–10 cm and 10–50 cm depths and at 10 of those points for 50–100 cm depth using a 25-mm diameter stainless steel Hoffer tube sampler and bulked for analysis at the plot level per depth. Two soil bulk density cores were collected for 0–10 cm and bulked per plot, and one soil core was collected for the deeper soil (10–20, 50–100 cm) at one random location within each plot using a 10 cm diameter × 10 cm length steel core. No soil water solution was sampled so no direct assessment of leaching potential could be reported.

All field fresh samples (LFH and soil) were kept cool before being analysed for total hexazinone concentration and total terbuthylazine concentration along with the concentration of the major metabolite of environmental concern terbuthylazine-desethyl using the same method described in Garrett et al. (2015). These compounds were extracted using ethyl acetate as the solvent and then analysed by gas chromatography/mass spectrometry (GCMS). The extraction method was a modified method described by Roos et al. (1987) with 80–90 % terbuthylazine and hexazinone recovery validated with samples of known concentration (RJ Hill Laboratories, Hamilton, New Zealand; http://www.hill-laboratories.com/). Detection limits were 0.001 mg kg^{-1} for mineral soil and a detection limit of 0.01–0.08 mg kg^{-1} for LFH. For the metabolite terbuthylazine-desethyl, the detection limit was 0.18–0.007 mg kg^{-1} for both mineral soil and LFH. Subsamples of the initial soil samples were air dried, sieved and the <2-mm soil fraction analysed for total organic carbon using a LECO FPS-2000 CNS thermal combustion furnace (modified Dumas method) and pH using 1:2.5 soil to water ratio. The soil bulk density samples were sieved to attain soil fractions (<2 and >2 mm) and then oven dried (105 °C) before being weighed. The LFH and woody debris components were dried separately at 70 °C before being weighed.

Data and statistical analysis

As described in Garrett et al. (2015), a daily water-balance model was used to calculate both root-zone water storage and drainage from the root zone extending to 1-m soil depth in order to assist with interpretation of active ingredient movement and potential of loss via leaching. Daily meteorological data required for the water-balance model were obtained from a meteorological station on-site and included total rainfall and solar radiation, mean air temperature, minimum air temperature and average vapour pressure deficit. The model was calibrated using measured volumetric soil water content, which showed a high correlation with modelled values (Fig. 1a).

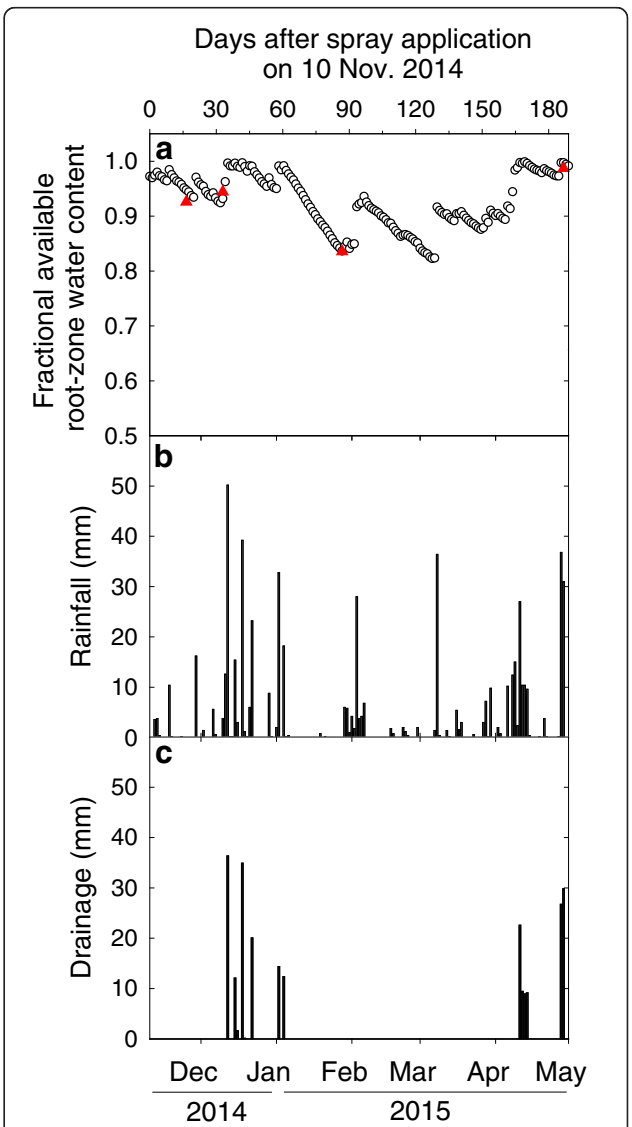

Fig. 1 Seasonal variation during the 2014/2015 monitoring period in (**a**) fractional available root-zone water content, **b** rainfall and **c** predicted drainage. **a** Modelled (*open circles*) and measured (*filled red triangles*) fractional available root-zone water content are shown

The total amount (stock) (kg ha^{-1}) of the active ingredients terbuthylazine, terbuthylazine-desethyl and hexazinone present in the LFH at each sampling time were calculated using the chemical concentration multiplied by the total oven dry mass of LFH over a known area. For the soil samples, chemical concentration was multiplied by the corresponding total bulk density and sample thickness. All reported stocks are expressed on a horizontal-area basis by correcting for plot slope.

All analyses were undertaken using the SAS software package (SAS Institute Inc. 2008). Total stocks of terbuthylazine and hexazinone for each plot at each measurement date were expressed as a percentage of the quantity recorded on the spray date. Changes in the percentage of each active ingredient, D, over time, t, were modelled using the following equation,

$$D = \left(\left(1-(1-\exp(\alpha t))^{\beta}\right)100\right) \quad (1)$$

where α and β are empirically determined parameters. Model precision was determined through examination of the root mean square error (RMSE) and coefficient of determination (R^2).

Findings

Rainfall and drainage

Rainfall was evenly dispersed in the first month after spray application and relatively low compared with the following 2 months (Fig. 1b), resulting in no predicted drainage within the first month after spray application (Fig. 1c). After the first month, there were six predicted drainage events ranging from 12 to 36 mm, which occurred during December and January (Fig. 1c). Little rain fell between February and early April (Fig. 1b), and no further drainage was predicted (Fig. 1c). Heavy rainfall during two events in April (Fig. 1b) resulted in total predicted drainage of 107 mm over this month (Fig. 1c).

Spray application

On the day of spray application, the weather was fine and there was a slight 5 km h^{-1} wind from a predominantly southern direction. The average application of Agpro Valzine Extra to the soil plots was 16.4 L ha^{-1} (se = 2.5 L ha^{-1}), which equates to an average rate of 7.3 kg ha^{-1} (se = 1.1 kg ha^{-1}) for terbuthylazine and 1.8 kg ha^{-1} (se = 0.3 kg ha^{-1}) for hexazinone.

Forest floor and soil

The average mass for coarse woody debris (>10 cm diameter) was 16.8 t ha^{-1} (se = 2.6 t ha^{-1}) and for LFH was 31.5 t ha^{-1} (se = 2.6 t ha^{-1}). Coarse woody debris and LFH covered 64 % (se = 3 %) of the ground area after harvesting activities, the remaining area constituted mineral soil. The soil carbon concentration, pH and bulk density are shown in Table 1.

No trace of either terbuthylazine or hexazinone was detected in the initial LFH and soil measurements taken before the 2014 spray application (data not shown). Both terbuthylazine and hexazinone concentrations declined rapidly with time following spray application (Fig. 2). Both active ingredients were mostly found in the top layer of soil (i.e. 0–10 cm), followed by the litter layer. Relatively little terbuthylazine or hexazinone was found at soil depths below 10 cm. An increase in hexazinone amount was recorded in the 10–50 cm soil layer 79 days after spray application (Fig. 2). Either no trace or a very low concentration of the metabolite terbuthylazine-desethyl was detected over the entire monitoring period in both the LFH and soil (data not shown).

The dissipation model (Eq. (1)) explained the data very well for both terbuthylazine and hexazinone (Fig. 3). Coefficients of determination for the model fitted to the replicate data, averaged 0.998 (range 0.996–1.000) for terbuthylazine and 0.999 (range 0.997–1.000) for hexazinone. The modelled root mean square error was relatively low, averaging 2.00 % (range 0.675–2.93 %) for terbuthylazine and 1.57 % (range 0.939–2.77 %) for hexazinone. For both herbicides, there was a steep decline in the percentage of active ingredient remaining with time following spray application (Fig. 3). Half-lives for terbuthylazine and hexazinone were, respectively, 0.059 days (se = 0.033 days) and 0.738 days (se = 0.426 days).

Discussion

The operational aerial application of terbuthylazine and hexazinone to a steep Recent Soil site after recent harvesting of *P. radiata* forest provided evidence that a proportion of both active ingredients were retained in the litter (Fig. 2). This is consistent with the results from a planted forest on Pumice Soil (Garrett et al. 2015) and demonstrates the importance of litter (LFH) and harvest residues in the retention of both these active ingredients

Table 1 Soil chemical and physical characteristics for the field experimental site. Values shown include the means followed by the standard error in brackets

Soil depth (cm)	pH	Total C[a] (%)	Fine earth (<2 mm) bulk density[b] (g cm^{-3})
0–10	5.6 (0.41)	5.4 (0.31)	0.52 (0.03)
10–50	5.3 (0.80)	2.6 (0.28)	0.52 (0.02)
50–100	4.9 (0.93)	0.8 (0.11)	0.75 (0.16)

[a]Fine earth fraction
[b]No soil coarse fraction, total bulk density equals fine earth fraction bulk density

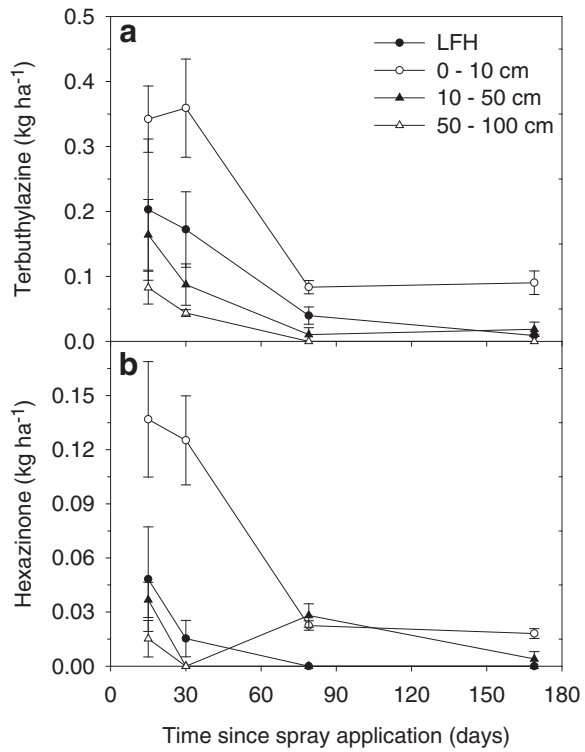

Fig. 2 Changes over time since spray application in the amount (kg ha^{-1}) of (**a**) terbuthylazine and (**b**) hexazinone, within the litter, LFH (*filled circles*) and at soil depths of 0–10 (*open circles*), 10–50 (*filled triangles*) and 50–100 cm (*open triangles*). Values shown include the mean and standard error

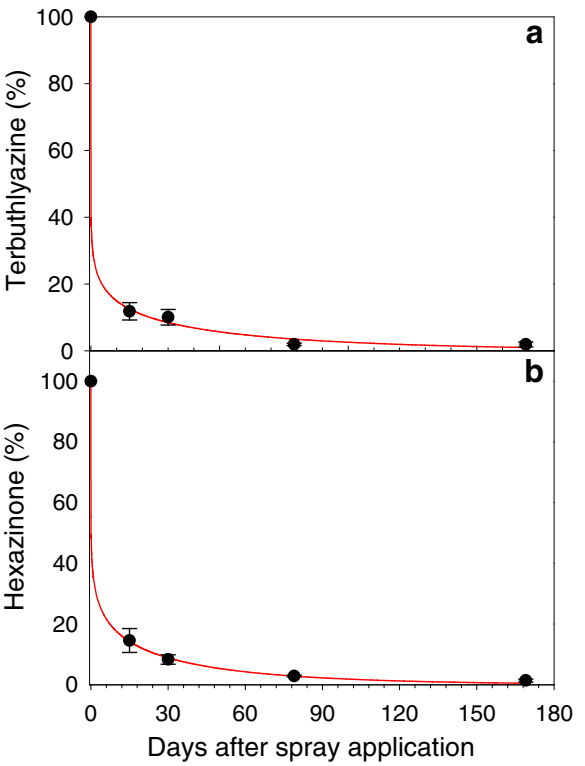

Fig. 3 Changes over time since spray application in the remaining percentage of (**a**) terbuthylazine and (**b**) hexazinone. Values shown include the modelled values (*solid red line*) and the mean and standard error of measurements (*filled black circles*)

on-site. No predicted drainage below 1-m soil depth occurred for either herbicide within the first month since there were no significant rainfall events during that period. This period was identified as the time of greatest potential risk of herbicide movement off-site by Garrett et al. (2015).

The amount of forest floor cover present at the site was lower (16.8 t ha^{-1}) than that at a Pumice Soil site (29.4 t ha^{-1}) studied by Garrett et al. (2015), so there was less organic matter on-site for retention of active ingredients. As a result of this difference, the retention of active ingredients occurred mostly in the top 10 cm of soil at the Recent Soil site (Fig. 2), whereas the majority of the retention was in the litter layer at the Pumice Soil site. The results from the current study do indicate that there was some limited movement of the active ingredients down the soil profile within the first month, most likely due to small rainfall events during this time resulting in water movement through the forest floor and upper soil surface layers (Fig. 2). Terbuthylazine was relatively immobile, with the active ingredient still detectable in the 0–10 cm soil layer and litter 79 days after spray application. These results are most likely due to the low water solubility and the high capacity for adsorption to organic carbon of this active ingredient (Tomlin 2006; Watt et al. 2010). In contrast, no hexazinone was detected in the litter 79 days after spray application. Some hexazinone was still present in the 0–10 cm soil, but the amount of this active ingredient increased in the 10–50 cm soil depth between 30 and 79 days after spray application (Fig. 2) largely as a result of a number of drainage events (Fig. 1). The first drainage event occurred 34 days after spray application after a 54 mm rainfall event (Fig. 1). Baillie (2016) measured an increase in the concentrations of both active ingredients in the sub-catchment stream water 2 days post this drainage event. The higher water solubility of hexazinone (Rolando and Watt 2012) compared with terbuthylazine, in particular, results in hexazinone having a higher potential to leach through the soil with drainage events. This is consistent with the results reported for a planted forest Pumice Soil and with the behaviour of hexazinone in soil from other studies (see Garrett et al. 2015 for a more detailed summary). It is worth noting that the Recent Soil at the site studied had a higher soil carbon concentration (Table 1) than other surveyed recent soils that underlay planted forests (5.4 % compared to

an average of 1.6 % soil organic carbon at depth of 0–10 cm). Therefore, the studied soil should have a higher capacity to retain terbuthylazine compared with the surveyed Recent Soils reported on by Watt et al. (2010).

The predicted half-lives of both terbuthylazine and hexazinone calculated in the current study were very short compared with previous estimates in New Zealand soils, including a similar study in planted forest Pumice Soil where half-lives for terbuthylazine and hexazinone were calculated as 10 and 18 days, respectively (Garrett et al. 2015). Environmental conditions and rates of application were very similar between this study and the site with Pumice Soil (Garrett et al. 2015), so it is unclear why there were higher rates of dissipation on the Recent Soil. Both active ingredients have been shown to provide effective residual weed control post-application, with Tran et al. (2015) reporting effects on broom seedling survival and growth in a silt loam soil for 3 to 9 months. Although the rapid dissipation of both active ingredients in the Recent Soil suggest a short period of environmental concern, it would be useful to determine if these low amounts can provide effective weed control over the longer term (6 months after spray application).

In this study, only 10 % of both active ingredients remained 15 days after spray application indicating that the first 2 weeks represented the greatest risk period for off-site movement. The low risk of off-site movement is supported by the associated research on the environmental fate of these two active ingredients in the aquatic environment (Baillie 2016). Further research is required to confirm the rapid dissipation rate of these active ingredients in Recent Soils at other sites and under different climatic conditions, and for other soil types that are dominant in New Zealand planted forests.

Conclusion

Aerially applied terbuthylazine and hexazinone was found to have the greatest potential to move off a steep Recent Soil site during the first 2 weeks following application. After this time, the likelihood of further movement will be low due to the very rapid dissipation of both active ingredients, plus their retention in the litter layer and 0–10 cm soil depth. The management of organic matter, including litter (LFH) and harvest residues, clearly plays an important role in determining the environmental fate of both terbuthylazine and hexazinone. Further research should be undertaken to confirm these findings.

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Authors' contributions

LG was the primary author and with SP conducted the majority of the field work. MW undertook the data analysis and assisted with the writing of the paper. All authors read and approved the final manuscript.

Competing interests

The authors declare that they have no competing interests.

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