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Chlorinated pesticides in stream sediments from organic, integrated and conventional farms

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ABSTRACT

To determine if current sheep/beef farming practices affect pesticide residues in streams, current-use and legacy chlorinated pesticides were quantified in 100 sediment samples from 15 streams on the South Island of New Zealand. The study involved five blocks of three neighboring farms, with each block containing farms managed by organic, integrated and conventional farming practices. Significantly higher concentrations of dieldrin, $\sum$ endosulfans, $\sum$ current-use pesticides, and $\sum$ chlorinated pesticides were measured in sediments from conventional farms compared to organic and integrated farms. However, streams in the latter two farming categories were not pesticide-free and sometimes contained relatively high concentrations of legacy pesticides. Comparison of measured pesticide concentrations with sediment quality guidelines showed that, regardless of farming practice, mean pesticide concentrations were below the recommended toxicity thresholds. However, up to 23% of individual samples contained chlorpyrifos, endosulfan sulfate, $\sum$ DDT, dieldrin, or $\sum$ chlordane concentrations above these thresholds.

1. Introduction

Scientists, regulators, farmers and the public from locations around the globe are increasingly interested in understanding how different farm management approaches (such as organic, integrated, and conventional) affect the environment (Bengtsson et al., 2005; Chatupote et al., 2005; Gomiero et al., 2011; Hole et al., 2005; Magbanua et al., 2010; Van Diepeningen et al., 2006). Organic farming uses environmentally friendly approaches for weed, pest, soil and disease management and avoids the use of synthetic pesticides and fertilizer (Šrůtek and Urban, 2008). In conventional farming, these chemicals are used and a generally more industrial approach to crop production is taken (Loake, 2001). In contrast, the integrated farming approach aims to achieve optimal results from both economic and environmental perspectives by encouraging beneficial predators of pest species and by using synthetic fertilizers and pesticides, but to a minimal degree (Nemecek et al., 2011).

The relative impacts of organic, integrated, and conventional sheep/beef farms on the physicochemical properties of streams (e.g. total dissolved nitrogen and fine sediment on the bed) and on stream macroinvertebrates were investigated by Magbanua et al. (2010), representing the first study to compare the effects of these three farming types on streams. In that study, glyphosate, one of the most widely used herbicides worldwide (Gasnier et al., 2009), was also quantified in stream sediments. Magbanua et al. (2010) reported that, while all three farming practices reduced stream health, conventional farming affected both stream physicochemistry and macroinvertebrate communities more than either organic or integrated farming practices. Similarly, glyphosate concentrations were significantly higher in stream sediment samples collected on conventional farms than in stream sediment from organic or integrated farms, leading the authors to propose that glyphosate concentrations may have contributed to the poorer biological health in streams on conventional farms. However, these authors recognized that other pesticides or agrochemicals may have also contributed to the observed results and the work presented herein was designed, in part, to address those questions.

Pesticides, especially chlorinated ones, have raised global concern due to their long persistence in the environment, impact

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on non-target organisms, and bioaccumulation in the tissue of animals and humans via the food chain (Chau, 2005; Tang et al., 2007). Until the 1970s, chlorinated pesticides were the most widely used pesticides in the world (Zhang et al., 2011). They can be introduced to the environment via several routes; however, agriculture is generally the most important (Arias et al., 2011; Miglioranza et al., 2004, 2002; Salvadó et al., 2006). Several studies have investigated pesticide concentrations in agricultural streams. For instance, the concentrations and distributions of chlorinated pesticides were reported in streambed sediments by Phillips et al. (2010) and in suspended fine sediment by Schäfer et al. (2008) and Hladik et al. (2009). Although pesticides have been measured in farmland streams before, to the best of our knowledge, no previous studies have compared pesticide concentrations (except for glyphosate in Magbanua et al. (2010), see above) in streams draining organic, integrated, and conventional farms.

The present study aimed to (1) measure the concentrations of current-use and legacy chlorinated pesticides in sediments from 15 farmland streams on the South Island of New Zealand, (2) test several initial hypotheses about the expected effects of different sheep/beef farming practices (organic, integrated, and conventional farming) on pesticide distributions, and (3) compare measured concentrations with stream quality guidelines. The significance of this work arises, in particular, from the initial hypotheses about pesticide distribution patterns being disproven, highlighting the complexity of pesticide behavior in the environment and the importance of both pesticide vapor drift and increased pesticide runoff at conventional farms.

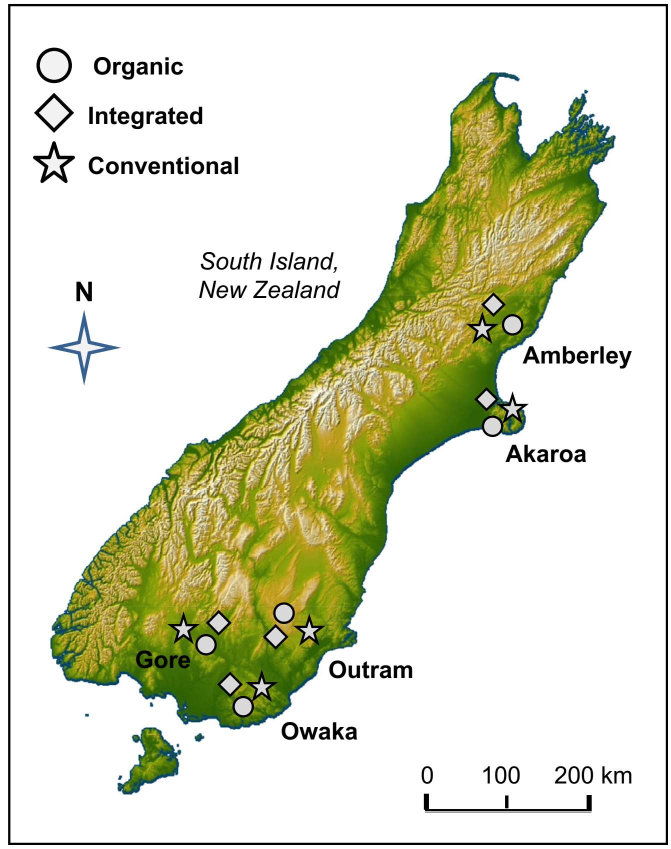

Fig. 1. Locations of stream sites where sediment samples were collected.

2. Materials and methods

Sources of chemicals, analytical standards, and other supplies are described in Section 1 of the Supplementary Information (SI).

2.1. Sampling sites

Sediment samples were collected from 15 streams passing through sheep/beef farms (the same streams as the ones sampled by Magbanua et al. (2010)) at the locations shown in Fig. 1. The farms were arranged in clusters at five locations near the towns of Amberley, Akaroa, Outram, Owaka, and Gore on the South Island of New Zealand. Detailed information about the five farm clusters is presented in Section 2 and Table S1 in the SI. Sediment sampling was conducted during late Austral spring/early summer, in November–December 2007. During spring and summer, farming activities increase, and this coincides with the period when farmland weeds and pests become most active, resulting in more intense application of pesticides compared to winter or autumn.

Each of the five clusters was comprised of three neighboring farms; each farm contained a stream and practiced sheep/beef farming. In each cluster, one farm was managed using organic, one using integrated, and one using conventional practices. The organic and integrated farms were converted to their current status between 8 and 11 years prior to this study being conducted (Magbanua et al., 2010). The average stream widths at organic, integrated, and conventional sites were 2.0 ± 0.9, 1.5 ± 0.3, 1.9 ± 0.6 (standard error, SE) m, respectively. Average current velocities were 13.4 ± 3.7, 24.2 ± 10.4, and 16.8 ± 10.6 (SE cm s^{-1}), respectively. At each stream, nine streambed sediment samples were collected; three were from near the upstream farm boundary (but still well within the property, averaging 244 ± 63 (SE) m downstream of the boundary), three were located midstream with respect to the property, and three were near the lower farm boundary (3809 ± 497 m downstream of the uppermost sites and 363 ± 124 m above the lower border). This design resulted in a total of 135 samples from 45 sites (see Section 3 in the SI for details of sampling protocol).

2.2. Analytical procedure

Out of 135 samples that were collected, only 100 contained a sufficient amount of sediment (>5 g) to proceed with the analysis; the other samples contained >90% gravel so could not be analyzed. Of the 35 samples that were not analyzed, 11, 5, 9, and 10 were from the Amberley, Akaroa, Outram, and Gore regions, respectively. Sediment samples were extracted using selective pressurized liquid extraction (S-PLE). In this S-PLE method, Florisil (magnesium silicate) was used to remove interfering compounds, such as lipids and biomolecules, originating from the sample matrix. This extraction method was based on that previously described by Hussen et al. (2007) for the analysis of organochlorine pesticides in soil samples (see Section 4 in the SI for more details).

Sample extracts were analyzed using an Agilent (Santa Clara CA, USA) gas chromatograph (GC 6890N) coupled with an Agilent mass selective detector (MS 5975B) (see Section 5 and Table S2A and S2B in the SI for details about the GC/MS analysis, data quantification, and quality control). The 19 legacy pesticides and degradation products on the target analyte list were α-hexachlorohexane (HCH), β-HCH, γ-HCH, δ-HCH, *o,p*′-dichlorodiphenyltrichloroethane (DDT), *p,p*′-DDT, *o,p*′-dichlorodiphenyldichloroethane (DDD), *p,p*′-DDD, *p,p*′-dichlorodiphenyldichloroethylene (DDE), aldrin, heptachlor, dieldrin, endrin, *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, endrin aldehyde, and heptachlor epoxide. The targeted current-use pesticides (i.e. registered for use in New Zealand when the sampling took place) were α-endosulfan, β-endosulfan, chlorpyrifos, and trifluralin; endosulfan sulfate, the degradation product of the endosulfan isomers, was also targeted. Analytical method detection limits were calculated based on USEPA method 8280A (USEPA, 1996). The individual detection limits ranged from 0.0002 to 0.5200 ng g^{-1} (see Table S3 in the SI for more details).

The current-use pesticides selected for analysis in this study have been widely applied on New Zealand farmlands (Chapman, 2010; Park et al., 2009), were previously detected in New Zealand groundwater systems (Close and Flintoft, 2004; Gaw et al., 2008; Sarmah et al., 2004), are highly toxic compared to most other current-use pesticides and are potential endocrine disruptors (Kegley et al., 2010), and have relatively high volatilization potentials (making them prone to undergo vapor drift between farms) (Davie-Martin et al., 2012). Limited information is available about the current and historic use of specific pesticides on the investigated farms; however, the Agriculture Research Group on Sustainability (ARGOS) at the University of Otago compiled a list of pesticides used on these farms during the 2005–06 summer (personal communication). Of our targeted current-use pesticides, only trifluralin, a herbicide used to control weeds in sheep/beef farms, was reported to have been used in 2005–06. Other pesticides, including acidic and sulfonylurea herbicides and other organophosphate insecticides, were reported to have been used; however, those chemicals were not targeted because the aim of this study was to measure the more toxic and persistent organochlorine pesticides. Glyphosate was widely used in the study areas and as previously mentioned, its concentrations have been reported by Magbanua et al. (2010).