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Evaluating and controlling pharmaceutical emissions from dairy farms: a critical first step in developing a preventative management approach

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ABSTRACT

Concern that pharmaceuticals may be escaping into the environment where the potency and persistence of certain compounds at trace concentrations could be chronically affecting biota is growing. Hitherto the main focus has been on human medications, personal care products and industrial endocrine disrupting chemicals. These generally enter sewerage systems where there is at least the prospect of partial removal by treatment plants before they enter waterways. By contrast, the agricultural sector, a significant user of veterinary pharmaceuticals, has no such treatment – compounds are deposited straight to ground in dung and urine or washed from hides in the case of topical applications.

This study investigates the fate of a number of antibiotic compounds (as well as the insect repellent, DEET, via a pilot assessment) used in herd health programs on dairy farms in the cow-rich Macalister Irrigation District in Victoria, Australia. Results from samples taken from irrigation drainage channels and streams demonstrate that these compounds *are* foot printing into an aquatic environment that extends to the Ramsar-designated Gippsland Lakes and associated wetland system. Conclusions are drawn as to how this problem might be lessened by a targeted water quality monitoring program and some rather straightforward changes to farm management practices.

1. Introduction

The discovery of pharmaceutically active chemicals in European, Japanese and North American groundwater, rivers and lakes in recent years has caused concern in regulatory and scientific circles [1]. Whereas humans and/or animals, for which drugs have been developed, may be exposed to a relatively constant and 'acceptable' range of blood/tissue concentrations for a defined period of treatment smaller organisms in the receiving environment can be chronically exposed at these concentrations. In other words their continual infusion into the aquatic environment serves to sustain perpetual life-cycle exposures for aquatic organisms [1].

A 'signature science' responsibility of the U.S. EPA's Office of Research and Development *is to pioneer and nurture new programs for identifying, evaluating, and developing the requisite science for minimising existing, or preventing future, exposure risks from previously unrecognized/unexpected chemicals*. One such set of unregulated chemicals are pharmaceuticals: release to the environment of these compounds especially when they are toxic and known to persist in metabolised or recombined states, is an issue of growing concern worldwide following their discovery in European, Japanese and North American groundwater, rivers and lakes [1–8]. Recent analytical tests on water in the Brisbane River, Australia and influent and effluent at a waste water treatment plant also in Brisbane as well as agricultural drains in East Gippsland, Australia [9–11] are among a number of local studies which have confirmed the presence of significant levels of antibiotics and the insect repellent DEET (*N, N*-diethyltoluamide).

Field studies of human drug release have indicated the sorts of impacts that could derive from various drugs [1]:

- Endocrine disruption – due to natural and synthetic oestrogens (contraceptive pill), thyroid growth regulators and baldness treatments.
- Hyper-sensitivity to pollutants by blocking transporter mechanisms that remove contaminants from cells of aquatic animals – due to blood pressure, immunosuppressive and heart drugs.
- Spawning boost in bivalves – due to anti-depressives.
- Change in bacterial flora of soil and groundwater – due to antibiotics.

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A combination of increased usage, greater awareness and developments in sample enrichment and analytical techniques, specifically liquid chromatography – tandem mass spectrometry (biochemical analysis) or LC–MSMS, has led to a wide range of investigations on pharmaceuticals in the environment. For example, the United States Geological Survey (USGS) has completed a reconnaissance of the occurrence of pharmaceuticals, hormones and other organic waste water contaminants in 139 American waterways [4]. In order to cover a wide variety of pharmaceuticals the study utilised five specialist laboratories and analysed for a total of 95 different analytes of which 85 were detectable in natural waters.

The most frequently detected compounds were coprostanol (fecal steroid), cholesterol (plant and animal steroid), DEET (insect repellent), caffeine (stimulant), triclosan (antimicrobial disinfectant), tri (2-chloroethyl) phosphate (fire retardant), and 4-nonylphenol (non-ionic detergent metabolite).

In the wake of this benchmark study Britain's Environment Agency has identified those compounds at greatest risk of exceeding "predicted no-effect concentrations" [3]. It has subsequently targeted 11 drugs, three of which are antibiotics, while implementing a monitoring program [12].

2. The agricultural issue

The Australian agricultural sector and the dairy industry in particular, are significant users of veterinary drugs, but there has been little study as to whether any of these compounds leach into and persist in local soil and aquatic ecosystems. Whereas the need to manage the environmental, human health and economic impacts of agricultural and veterinary (ag–vet) chemical use is emphasised in documents such as *Management of Agricultural and Veterinary Chemicals: A National Strategy* [13], there has been no attempt to develop specific management responses at an industry level. Most attention directed at pharmaceutically active chemicals has so far been focussed on their degradation within laboratories and at, or near, sewerage treatment plant outfalls where there is at least the prospect of some diminution via preceding treatment processes [10].

Pharmaceuticals routinely used in herd health programs include hormones, antibiotics, udder-cleaning antiseptics, anthelmintics and ecto-parasitic topicals. The extent that the active ingredients of any of these drugs (and their metabolites) leach into pastures, soil, runoff and groundwater is a matter for field research. In such circumstances, it is not known whether they react with each other as well as pesticides and herbicides forming further compounds which, either acting individually or in combination, could adversely affect bacteria, fungi and higher organisms [14].

3. This study

This study represents a key attempt to evaluate selected pharmaceuticals in Australian water bodies that receive potential emissions from dairy farms – in particular, the drains and streams in the Macalister Irrigation District (MID) situated in Gippsland, Victoria, Australia (Fig. 1). These waterways lie upstream from the Ramsar-designated Gippsland Lakes and their adjoining wetlands. MID farms have 300–800 cows per property representing one of the tightest packing of dairy herds in Australia short of feedlot production. The dairy farms are both flood and spray irrigated using water from the Glenmaggie and Cowwarr Weirs. The actual source of water supply to the MID is the Macalister, Thomson and Latrobe Rivers. The weirs are purely distribution structures.

According to an earlier CSIRO investigation [15] the Lakes and wetlands are at a cusp insofar as their survival is concerned. Further upstream, a report by the Co-operative Research Centre for Freshwater Ecology [16] highlighted the connection between farming practices and watercourse damage. It found…*a significant deterioration in the physical condition of the Thomson and Macalister channels, deterioration in both the in-stream and riparian habitat, and poor water quality. These, together with the major changes in the flow regimes, have had considerable impact on the ecology of these rivers, shown particularly by the reduced 'health' of the macro-invertebrate and fish communities. Additionally, the floodplain and associated wetlands* have *been largely cut off from the river channels through the building of levees*. However, in many cases the farmers did not build levees; they "enhanced" the natural levees by filling any low spots.

The rationale of the current study is to assist the conservation of the Gippsland Lakes and associated wetland system as a unique environmental, recreational and fishery resource by limiting the possibility of further damage to their very fragile ecology by more subtle water-borne contaminants. In particular, it aims to characterise the extent that ag–vet chemicals are present in the water bodies of the Latrobe River system which drains into the Morass Wetlands and Lake Wellington thence through The Straits into Lake Victoria.

In consequence, it seeks to increase awareness of the possible foot printing of such chemicals into aquatic ecosystems [17], the need for an on-going monitoring program, and the identification of basic control measures leading to the development of a leading edge practice regime for the dairy industry across Australia that would serve to regulate such emissions in contrast to the limited approach currently being followed [11].

3.1. Pilot assessment

Pursuant to a fuller sampling and analysis, a pilot study was undertaken during November 2002 to establish sampling venues, techniques and appropriate analytical methods [11].

In this pilot stage grab samples were taken from two channels draining a very dense network of dairy farms in the MID as well as a creek in the adjoining Snowy River system (Table 1). Each sample was collected in a 1-L glass bottle affixed to the end of a long pole. Exacting protocols were followed by the sampler to prevent contamination occurring with no wearing of insect repellents and careful rinsing of the sample bottles. Samples were refrigerated and sent to the National Research Centre for Environmental Toxicology (Entox) in Brisbane, Australia, where they were filtered and passed through an OASIS Hydrophilic – Lipophyllic – Balance (HLB) cartridge (60 mg). Cartridges were extracted and the eluate analysed by the Australian Government Analytical Laboratories according to techniques described by Kolpin et al. [4].

3.1.1. Results

The results of tests conducted on the three samples are shown in Table 2. Analytes were antibiotics and the insect repellent, DEET.

3.1.2. Analysis

Quantifiable concentrations of the insect repellent, DEET, were found in all three samples. This is in line with the USGS results which found that DEET was one of the most ubiquitous contaminants in American streams. No antibiotics were detected but Kolpin et al. [4] suggest that they could be absorbed by sediments over a long standing period and/or selected OWCs may be degrading into new, more persistent compounds instead of (or in addition to) their associated parent compound.

Of note was the severe drought conditions experienced in this region and throughout Australia in late November–early December 2002 and prior to sampling which may have influenced the results. It was thought at the time that the polar (water soluble) nature of many of the chemicals of interest, specifically monesin and enrofloxacin (see below), locked up on land could, following rainfall, be washed into nearby waterways possibly resulting in a different situation. However, later results reported below show