Pesticides: what the past teaches us

Auteurs :
SABATIER Pierre, Maître de conférences, Laboratoire EDYTEM (Environnement DYnamique et TErritoire de Montagne), Université Savoie Mont Blanc
POULENARD Jérôme, Professeur, Laboratoire EDYTEM (Environnement DYnamique et TErritoire de Montagne), Université Savoie Mont Blanc
ARNAUD Fabien, Directeur de Recherche CNRS, Laboratoire EDYTEM (Environnement DYnamique et TErritoire de Montagne), Université Savoie Mont Blanc

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analysis of lake sediments of an Alpine lake, covering a period of about 100 years, made it possible to follow the history of the use of herbicides, fungicides, insecticides and various treatments in a Savoy wine basin. This makes it possible to accurately track the history of chemical use, from the time they appear to the time they are officially banned. This study showed that the use of herbicides such as glyphosate, by removing the vegetation cover, has favoured soil erosion in vineyards and has led to the release of banned pesticides, such as DDT, which had remained stored in the soil of vineyards for many years after their prohibition and cessation of their use. These results indicate that the dynamics of pesticide storage in the environment, which is crucial in the assessment of ecotoxicological risks, must take into account possible future environmental disturbances on pesticide storage.

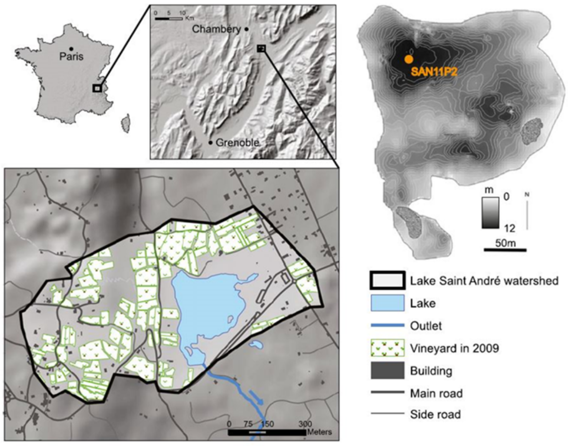

Figure 1. Lake Saint André and the surrounding vineyards (in 2009). The bathymetric map (topographic map of the dulac bottom showing depths by contour lines) shows the sampling site (SAN11P2) in the deepest (darkest on the map) area of the lake. [Source: Adapted from Sabatier et al.[2]]

The

Figure 2. The vines of the Lake of Saint André watershed. These vines were treated throughout the 20th century. [Source: © Pierre Sabatier]

France is the leading pesticide consuming country in Europe. The vine, representing 3% of national agricultural land but consuming about 15% of the pesticides marketed, is emblematic of this operation [1]. It is therefore logical to ask the question of the long-term effect of pesticide use in agriculture over the last century, for example through consequences on the environment and ecosystems. Currently, few studies have addressed this issue, mainly due to a lack of time lag in *in situ* measurements. An approach based on retro-observation of the environment has made it possible to fill this gap: lake sediment cores have been used to reconstruct the mobility dynamics of pesticides applied to vineyard plots in Savoie (Figures 1 & 2) over the past century [2].

1. Analysis of sediments in Lake St-André (Savoie)

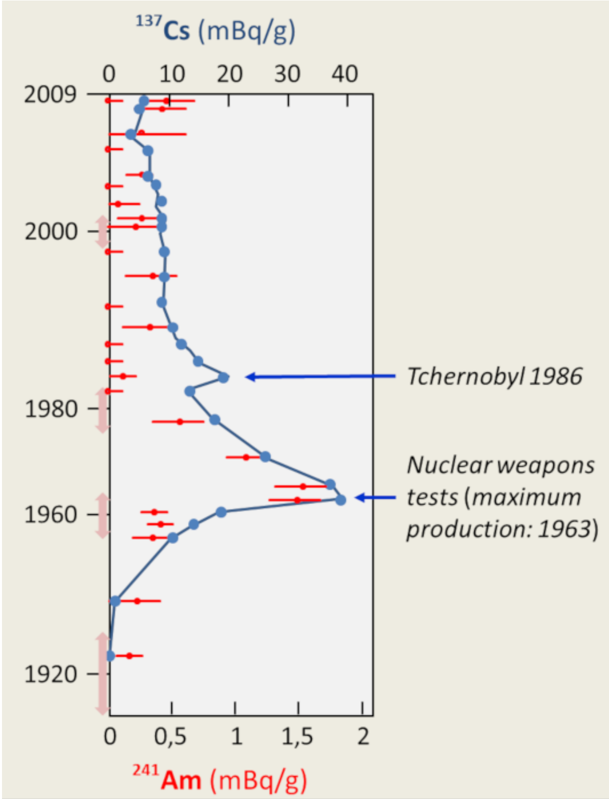

Figure 3. Distribution of Cesium 137 (137Cs) in the sediments of Lac St-André since 1920. The estimated age of the sediments (pink arrows) makes it possible to date the two peaks of 137Cs. The most important peak corresponds to the presence of 137Cs in the atmosphere due to nuclear tests (first fallout from 1955 and maximum of 137Cs in the atmosphere observed in 1963). 137Cs has gradually been deposited all over the planet (mostly in the northern hemisphere) These data are correlated with the presence of241Am (Americium, in red), an element from the disintegration of 241Pu (Plutonium) following nuclear tests. More recently, a significant but transient increase in 137Cs content has been observed in lake sediments following the Chernobyl disaster (1986). [Source: Adapted from Sabatier et al (see ref.[2])]

Sediment cores were collected from Lake Saint André (10 km south of Chambéry, see Figure 1) in 2011. These cores were then the subject of a multi-tracer study combining sedimentological and geochemical analyses. These analyses made it possible to characterize both the different sources contributing to the sedimentary filling of the lake, but also to measure the quantities of metallic trace elements (Copper, Lead...) and organic molecules, active substances in pesticides. Among all the molecules measured, twelve pesticides mainly attracted the attention of researchers. They are found more or less deeply buried depending on the years in which they were spread in the surrounding vineyards. These pesticides are classified into three broad categories and correspond to three herbicides, five fungicides and four insecticides. In parallel, sediment dating could be carried out using radioelements with short disintegration periods in order to obtain a time scale covering the last century. Natural radioelements such as ^{210Pb}, which has a half-life of 22.3 years, and artificial radioelements such as ^{137Cs} related to the fallout from the Chernobyl accident (1986) [3] or at most nuclear tests (1963) [4], have been measured (Figure 3). Thus, the first 45 centimetres of the cores studied could be dated and cover the last 120 years with two sudden changes in the sedimentation rate in the 1970s and 1990s, presenting for these two periods a doubling of the inputs of terrigenous material (material derived from surrounding soils) from the catchment area. Once the chronology was established, it was possible to study the presence of the different pesticides used in the treatment of the vine according to the period.

2. Bordeaux mixture and other fungicides

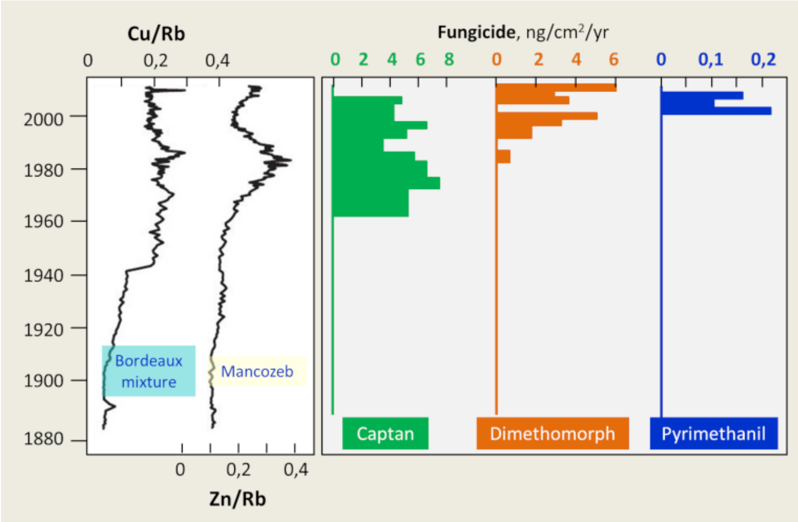

Figure 4. Distribution of fungicides in the sediments of Lake St-André since 1880. Bordeaux mixture and Mancozeb are compositions based on Copper and Zinc, respectively. Their quantity is estimated by comparison with the Rubidium content of the sediments, which is naturally present in the watershed. We can see that the use of Bordeaux mixture increased sharply after the Second World War. The right-hand side shows the distribution of synthetic fungicides and the successive appearances of Captan, Dimethomorph and Pyrimethanil in sediments can be monitored, following cultural practices. [Source: Adapted from Sabatier et al (see ref.[2])]

The first pesticide to be used and whose presence has been identified is the Bordeaux mixture, invented in Bordeaux at the end of the 19th century, made up of a mixture of copper sulphate and lime. It appeared in the recording at the beginning of the 20th century and showed a sharp increase at the end of the Second World War (Figure 4). It is a powerful fungicide used to control vine diseases such as powdery mildew and downy mildew. Other fungicides used to control these same diseases, such as Captan and Dimetomorph introduced later by agrochemical companies, are identified respectively from the 1950s and 1990s and follow one another over time (Figure 4). Some fungicides are also used in wine-growing to fight lesser-known diseases such as black rot, such as Mancozeb (based on Zinc) introduced in the 1960s and against grey rot with Pyrimethanil present from the 1990s onwards. The use of all these fungicides has been validated using the history of Savoyard vineyard practices; thus, the dates of appearance and disappearance are historically constrained and totally consistent with the reconstructions made from measurements on sediment cores.

3. Herbicides

For herbicides, three substances could be identified and quantified over time (Figure 5). First, in the 1960s, Atrazine degradation products accumulated in the sediment. This powerful herbicide was used in the late 1950s and banned in 2003. Atrazine was replaced by Glyphosate in the 1990s with the identification of AMPA, its metabolite. It is found up to the surface sediment. Glyphosate, still allowed in agriculture but much debated, is the active ingredient in Monsanto's Roundup®. It is used as a post-emergence, non-selective herbicide, widely marketed since the 1990s and highly effective in eradicating vegetation growing between rows of vines, leaving the soil bare. Finally, for the past 10 years, Diflufenican (introduced in the 1990s) has been present in lake sediments [2].

4. Insecticides

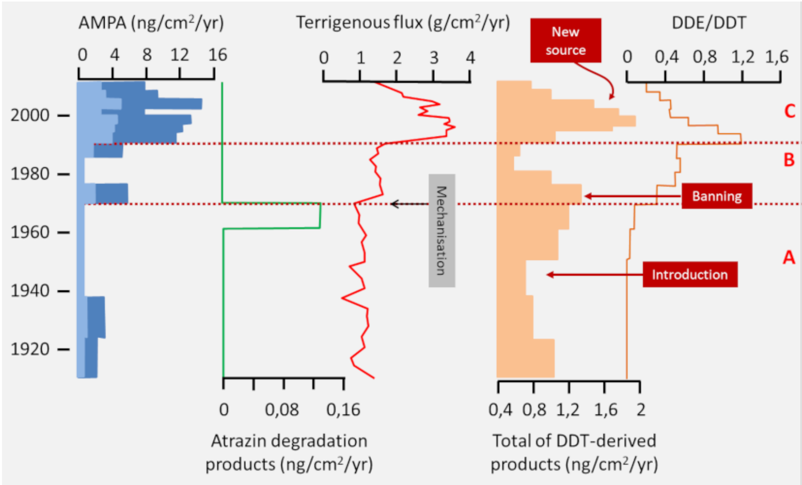

Figure 5. Impact of herbicide use on the nature of sediments in Lac St-André. The flow of terrifying residues (from the soils of the vineyards surrounding the lake) increased significantly in the 1970s and again after 1990. The first episode (A/B interface) is probably the combined result of the use of Atrazine on the one hand and the increase in mechanization on the other. The second phase (B/C interface) is directly related to the development of the use of Glyphosate (whose degradation product is AMPA; in light blue: detection limit). Plants growing around vines are destroyed by the herbicide, leaving the bare soil very susceptible to erosion. The amount of DDT (and DDE, which is a degradation product formed in the presence of oxygen) - an insecticide banned from use since 1972 - increased significantly in the mid-1990s. This increase in the accumulation of terrestrial products and DDT derivatives is most likely due to leaching during storms from weedy and therefore bare soils. The high values of the DDE/DDT ratio are related to the fact that DDT was degraded to DDE long after its use, so it has been released from soils that are easily erodible because they lack vegetation. [Source: Adapted from Sabatier et al (see ref.[2])]

Four insecticides have also been found in the sediments of Lake St. Andrew, mainly used to control crop pests. It was thus possible to highlight the succession over time of different molecules (Dicofol, Bromopropylate and Bifenthrin) according to the years of introduction and prohibition of these substances (Figure 5). This has been confirmed by surveys conducted with winegrowers. DDT (dichlorodiphenyltrichloroethane), a potent insecticide, first used for agricultural purposes after the Second World War, is also found in lake sediments from that period with a first peak concentration in the 1970s, just before its prohibition in France in 1972. The amount of DDT entering the lake decreases, but does not disappear, due to the high persistence of this molecule in the environment. DDT was recognized as highly toxic and very stable in the environment, and was banned in 1972 for health reasons. But, surprisingly, a second peak of DDT, significantly higher than the first, appears in sediments more than 20 years after its ban. Why is it that after a decrease in sediments in the 1980s, DDT is still present at high concentrations in recently deposited sediments? By looking at the metabolites of DDT [5] which are DDE (aerobically formed product) and DDD (anaerobically formed product) it is possible to highlight the origin of this new source of DDT found since the 1990s. It is actually DDT already present in the environment, partly degraded under aerobic conditions and re-mobilized from that period onwards; rather than a new introduction into the environment.

Therefore, the most likely hypothesis is that the massive use of herbicides in vineyards in the 1970s (Atrazine) and 1990s (Glyphosate) resulted in an increase in soil erosion of soils that were bare, and therefore easily erodible during rainfall, especially storms. These wine-growing soils, a true memory of past practices, contain high levels of pesticides and in particular DDT that is very stable in the environment. This DDT is thus remobilized through erosion processes resulting from the disappearance of vegetation between rows of vines due to the use of herbicides. This increase in soil erosion is also responsible for a greater accumulation of the amount of sediment transported and deposited in the lake, thus explaining the increases in the observed sedimentation rate but also leading to a significant loss of land for winegrowers. The first increase in erosion in the 1970s can also be linked to the significant increase in the mechanisation of wine-growing practices from that period onwards.

5. An environmental archive for the future

Since the 1950s, France has based its agricultural model on a massive use of plant protection products, becoming the third largest consumer of pesticides in the world [1]. Today, the impacts of massive contamination on professional users are known, but the effects of chronic contamination or exposure to pesticide cocktails are not yet well understood. This study shows that these molecules persist in our environment long after their bans, that they accumulate or are remobilized, depending on agricultural practices, and that even today, some very toxic molecules such as DDT, despite their ban for more than 40 years, remain present in our environment and can reappear to question us about the ever-increasing use of plant protection products. However, in recent years, the grassing of the rows of vines, encouraged by the Chamber of Agriculture, has significantly reduced the erosion of the vineyard plots reconstituted from lake sediments. In the coming years, emphasis will be placed on understanding the effect of these substances on organisms present in both the lake (fish, zooplankton, benthic macrofauna) and the watershed, using analyses of fossil DNA preserved in sediments, again based on the concept of retroobservation.

Through this study, it was therefore possible for the first time to carry out a sedimentary chronicle of pesticide use, which was validated by the history of the succession of chemical treatments of the vine in relation to the periods of introduction and prohibition. It also demonstrates the importance and need to take into account the long-term effects of pesticides in order to better assess the eco-toxicological risks associated with their use, particularly under conditions of environmental change. Finally, we can ask ourselves the question of the generalization of the processes highlighted here to other regions of France and the world, and to other types of crops because these pesticides, DDT, Glyphosate and others have been and are still used in most industrialized agriculture on a global scale.

References and notes

Cover image. Spreader suitable for spreading on vines. [Source: Karl Bauer own work (Original text: eigenes Foto) [CC BY 3.0 at], via Wikimedia Commons]

[1] Source INRA (2006 data, page 11): http://agriculture.gouv.fr/IMG/pdf/V3TAPButault7a26.pdf

[2] Sabatier P. *et al.* (2014) *Long-term relationships among pesticide applications, mobility, and soil erosion in a vineyard watershed*. PNAS 111, 15647-15652.

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