Chapter 1

Efficiency of Pesticide Alternatives in Non-Agricultural Areas

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http://dx.doi.org/10.5772/57150

1. Introduction

Global pesticide use is increasing, and such growth is recognized as stemming from agricultural needs in response to global food stress. However, pesticides are used on other areas than agricultural fields. Even if agricultural consumption of pesticides is undoubtedly the main use, the transfer from other, more impervious surfaces is regarded as a key point in understanding the fate and the global impact of pesticides, named biocides, when used for nonagricultural purposes. In the overall environment these chemicals are combined with those applied to agricultural areas, leading to confusion and thus a probable underestimation of nonagricultural pesticides. Numerous information campaigns have targeted agricultural users. The high remaining level of background contamination of rivers highlights that minoring even obliterating urban consumers precisely stultify the considered information campaigns. The ambiguous situation of port contamination will also be discussed in the present chapter.

However, nonagricultural uses mainly involve the same chemicals (e.g., herbicides) as agricultural uses. In the present chapter, the main biocides used will be listed, and then the differences in consumption depending on countries and legislation. The environmental traces of the main pesticides will be summarized with the confounding uses for watershed scale interpretation. The consequences of pesticide use depend on transfer rates, themselves conditioned by the type of surface where these chemicals are applied and their imperviousness. For highly artificialized urban areas, where biocides are mainly used, such information is pivotal: they explain a minor but significant part of aquatic environment contamination.

Alternatives to pesticide uses have been developed for decades, some even before the advent of pesticides, primarily herbicides. The present chapter will detail the alternative types, the respective efficiency depending on substratum and vegetation type. The discussion on the shortcomings of each alternative, the development level and the risks for humans or resulting from hazardous techniques (for both the environment and substratum) will distinguish promising techniques from those that have shown to be inapplicable. The authors will explain why technological impasses are patent and the possible ways to improve such technology to make them applicable. The mechanical techniques studied are mowing, brushing, rotative clogs, sweeping, and harrowing. Thermal techniques include solarization, high-pressure steam, foam mix, gas flames, infrared and to a larger extent, laser, electrocution and UV, microwave, and γ-radiation.

2. A semantic obstacle: How should pesticides and biocides be distinguished?

Far from being trivial, this question needs to be raised prior to examining international data. Indeed, if pesticides are used for crop protection, biocides are pesticide chemicals, i.e., poisonous compounds targeting pests, but non-agricultural pests. Incidentally, molecules involved and prophylactic control molecules could be the same.

Concerning the urban context, where the excessive use of pesticides is complicated by population density and the use of pesticides by local authorities is the most aggressive which targets healthy and ostentation goals both. Pesticide inputs could combine the two environmental contamination pathways. Considering urban use except for prophylactic campaigns, pesticides are spread in kitchen gardens and around ornamentals, i.e., gardens, golf courses, parks, including plant protection. However, use of the same chemical on roads and railways, facade building protection against termites, domestic and veterinary pest control all use biocides. In the present chapter "pesticide" will be used indifferently for pesticides and biocides, unless otherwise specified.

Use of sodium chlorate and iron sulphate should be precisely evaluated and taken into account: their amount is fourfold greater than non-mineral pesticides and could explain the discrepancy of the quantities reported. As for aquatic environment contamination, this controversy seems pointless: whatever the source, the chemical impact is the sole pragmatic yardstick.

3. Pesticide use

3.1. The pesticides used

First of all, inquiries on sales and applications of biocides should be considered carefully: some herbicides are indicated as being found in the urban watershed but could result from non-agricultural pesticide use in urban areas. For example, Gerecke *et al.* (2002), Devault *et al.* (2007), Gilliom (2007), and Botta *et al.* (2012) mention atrazine as consistently polluting the urban watershed without any homologation as a biocide.

Whatever the misuses, *glyphosate* and *diuron* are the most widely applied pesticides worldwide for biocide use. The main use of biocides is for weed control in developed countries, so glyphosate, whatever the geographical region, accounts for about half of non-agricultural use in terms of quantity, one-eighth of glyphosate sales (Hanke *et al.,* 2010; Blanchoud *et al.* 2007). Glyphosate is also used for agricultural ways to sign urban input, despite its increasing urban use. This trend is due to the ban of biocide use such as diuron (Okamura *et al.*, 2003; Gilliom, 2007), despite several marginal agricultural uses (vineyards and sugarcane in Australia (Haynes *et al.*, 2000)). Although diuron is banned for outdoor applications, it is found in veterinary devices and in antifouling paint additives (Irgarol 1051®), an emerging concern because of first-order kinetic façade leaching process (Burkhardt *et al*., 2011; Wittmer *et al*., 2011b). Even if Coutu *et al*. (2012) proposed a model integrating rainfall conditions depending on façade exposure, other climatic events (the effects of frost or sun) on building coatings and additives are not currently studied when examining the fate of façade chemicals. Non-agricultural use is reported to be equal or dominant for diuron (Bucheli *et al*., 1998; Gerecke *et al*., 2002; Wittmer *et al*., 2011).

Aminotriazole (also shortened to *amitrole*) for Europe (Blanchoud *et al.* 2004 and 2007) and *prometon* for the United States (Kimbrough & Litke, 1996; Bruce & McMahon, 1996; Hoffman *et al*., 2000; Philips & Bode, 2002, 2004, Ryberg *et al*., 2010) are respectively the third most referenced chemical in urban areas and marginal or even ignored in the US. For Ryberg et al. (2010), prometon use may be the most widespread biocide in the US Northeast and Midwest.

In the US, urban pesticide use seems to involve other pesticides than in Europe. Braman *et al*. (1997) noted the substantial use of pendimethalin (41%) in their study area near Atlanta but no diuron or amitrole, as Glozier *et al*. (2012) also noted. Amitrole and pendimethalin are not mentioned as urban pesticides contaminating US streams by Gilliom (2007). Even if pendimethalin is known in the European market, residues of pendimethalin are not indicated in European monitoring. Similarly, prometon is indicated (Kimbrough & Litke, 1996; Bruce & McMahon, 1996; Hoffman *et al*., 2000; Philips & Bode, 2002, 2004; Ryberg *et al*., 2010) as a major herbicide used in the urban environment. Philips & Bode (2004) highlighted that prometon concentrations in rivers were proportionate to the population density in the corresponding watershed, Gilliom (2007) listed it as the most frequently detected of the seven herbicides used in urban areas, and Ryberg *et al*. (2010) reported that, although prometon contamination was dominant in the US Northeast and Midwest, it was the most homogeneously represented herbicide for urban areas in the country as a whole. It is the most commonly used soil sterilant in urban areas (Kimbrough & Litke); locally unavailable to homeowners, it continues to be used in these areas by licensed applicators (*Ifid*.).

Only Gerecke *et al.* (2002) mentioned DEET (insect repellent) and diazinon (used by individual gardeners). Gilliom (2007), summarizing pesticide data in US streams and groundwater, showing that, between the six most relevant insecticides, five are significantly more frequently detected in urban streams than in agricultural streams. Four of them are significantly more detected in urban areas than in agricultural areas: diazinon, carbaryl, chlorpyrifos and malathion, despite substantial climate, biocoenosis and legacy diversities. Kimbrough & Litke (1996) already highlighted that urban insecticide use was greater and less diversified than agricultural insecticide use. Consequently, urban streams are more contaminated by such chemicals than agricultural streams. Diazinon, carbaryl, chlorpyrifos and malathion were the only insecticides used in urban areas and were noted by Whitmore *et al*. (1992) in the top 10% of the most frequently used pesticides by homeowners and certified applicators out of 312 compounds identified.

In the Croton Lake watershed, near New York City, Philips & Bode (2002 and 2004) also inventoried diazinon, in addition to carbaryl and imidacloprid. However, diazinon (with prometon) is the only one indicated as being present in densely populated watersheds (Philips & Bode, 2002, 2004). In a Californian urban context, Walters *et al*. noted carbaryl contamination due to *Homalodisca coagulata* (Say) infestation.

For urban use, Moran (2010) and Jiang & Gan (2012) reported that, in California, pyrethroids are the most widely used pesticides for urban areas. Weston & Lydy (2012) focused on pyrethroids due to their representativeness.

Considering "urban streams" as coming from watersheds whose land use was at least 25% urban and at the most 25% agricultural, Ryberg *et al*. (2010) listed pesticide residues and trends: if prometon was the herbicide the most frequently found in US rivers, herbicide trends are described as mixed. Although s-triazines are the main monitored pesticides, simazine and atrazine are more often found in rural areas. Neither a downward nor an upward trend seems to dominate, even if atrazine metabolite DEA is increasing compared to active chemicals.

3.2. Quantities

Wittmer *et al*. (2011a) noted that urban biocide consumption is within the same range as agricultural pesticides in Switzerland (1300–2000 t each), like Pissard *et al*. (2005) in Belgium. Similarly, Lassen *et al*. (2001) conclude that Denmark has high biocide consumption. This question is pivotal and could explain the clear differences between authors: Blanchoud *et al*. (2004) consider nonagricultural chemicals as approximately 1% of the total amount in the Marne River watershed (France), in accordance with several authors (Chauvel, pers. comm.), whereas Aspelin (1998) estimates it at about 25% for the US and previous authors (Lassen *et al*., 2001; BLW, 2007; FriedliPartner *et al*. 2007) at about 50%.

Approximately 10% of pesticide quantities spread stem from nonagricultural use in developed countries (Hanke *et al*., 2010; Kristoffersen *et al*., 2008). Municipalities maintain recreational gardens and playing fields. Even if athletes and the young are more exposed in such places (Harris & Solomon, 1992; Bernard *et al*., 2001; Chaigneau, 2004), this contamination pathway is not identified as a major one. Sports fields are roughly counted because of their heterogeneity: villages could present turf areas as a sports field that cannot be compared with large cities' equipment. That said, about 30,000 sports grounds have been inventoried in France: about one per town, as in all developed countries. Amenity use accounted for approximately 0.19% of pesticide use in Denmark, about 2.7% for the Netherlands and the United Kingdom, less than 3.4% for Germany, 0.6% for Finland and 1% in France (Blanchoud *et al*., 2004).

These results, considering minor surfaces with regard to local and even global land use, is due to greater urban use of pesticides, in comparison to the same surface treated, than in rural areas (Barbash & Resek, 1996; Devault, 2007). Barbash & Resek (1998) considered that lawns received 7.4 kg/ha (insecticide: 2.4 kg/ha; herbicide: 5 kg/ha), golf courses 18.8 kg/ha (insecticide: 13 kg/

ha; herbicide: 5.8 kg/ha), whereas agricultural areas received 2.3 kg/ha (0.9 kg/ha herbicide and 1.4 kg/ha insecticide).

It is also valuable to compare these results, from survey questionnaires, completed on a volunteer basis with the estimation from Aspelin (1997) and UIPP (2000): even if pesticide use is tending to decrease, other biases should be put forward: (1) hidden pesticide use such as flea collars), (2) the spontaneous trend to minimize one's own pesticide use, and (3) the lack of pesticide traceability.

3.2.1. Trends in developing countries

Developing countries' urban areas form a related context (Ecobichon, 2001), the subject of increasing concern. To provide the least expensive off-season fresh fruit (Forget *et al*., 1993), more acutely toxic and persistent pesticides are used in developing countries (Schaefers, 1996). The trend is similar for biocide use of pesticides: pyrethroid esters are used for household spraying to repel or kill tropical disease vectors (mainly biting insects), which are nine times more expensive than DDT (Webster, 2000): without international sponsoring, poorer nations often limit or abandon control programs. Older but restricted pesticides are not patented: local or regional chemical synthesis could occur because international bans are not applied, despite the Stockholm and Basel conventions. Thus, the main pesticide intoxications occur in developing countries: data are biased by unreported cases, but the World Health Organization reported 3 million severe poisonings, including suicides and 220,000 deaths for 1990. Such results, which have since been corroborated, are caused by careless handling and home storage (under beds, on kitchen shelves (Ecobichon, 2001), lack of protective equipment (possibly due to discomfort), and individual, collective or governmental actions (Gomes *et al*., 1999), consumption of food or beverages stored in pesticide containers for improper uses (water or food storage). Commuters may produce food in kitchen gardens but male handwork is mainly employed in cash-paying jobs in plantations surrounding cities or in industries: once planting has been completed, crop care is in the hands of women and older children, along with child care and domestic tasks. These tasks induce frequent comings and goings between indoors and the garden, enhancing pesticide exposure risks. Kitchen garden care and maintenance is so devoted to inattentive and overbusied female or infantile handwork As in developed countries, but more acutely, the long-term solution to pesticide problems is education (Ecobichon, 2000), but developing countries lack the regulatory framework, due to insufficient awareness, means and trained personnel for these controls (Ecobichon, 2001).

In all countries, more than 50% of private gardens are treated with pesticides; Hanke *et al*. (2010); in Switzerland the percentage is estimated at 90%, 60% are total herbicides for terraces and about 30% are selective herbicides for grass, shrubs and trees. Fungicides, insecticides and other pesticides (against rodents, mollusks, etc.) account for about 4% each. Thus, the main individual consumption is for esthetics, not for kitchen gardens. Twenty percent of the Swiss population spread pesticides on walkways and garden paths, although this is strictly forbidden in Switzerland (Hanke *et al*., 2010).

In France, where about 1,100,000 ha are grassed, 605,000 ha are residential, including 23,000 ha of apartment buildings: gardens remain a status symbol. Consequently, the main grassed surface is under private control without adequate training, subjected to unclear application protocols, and receive about 5000 t of pesticides every year. For example, park treatment information is conventionally provided for a 600-m² applications, due to large rural gardens and parks: the indicated quantity to use could be scrupulously determined but is often interpreted incorrectly: many users only spread pesticides on a limited surface, i.e., a few square meters, but use the dose for 600 m² because they do not understand the instructions for use. This information base could also be lacking because 20% of individual gardeners say they are unaware of the impact of pesticides on health and the environment (French Ministry of Ecology, 2011).

In the US, Voss *et al*. (1999) identified diazinon, 2-4D, and mecoprop as the main pesticides polluting streams during rainstorms and successfully compared them to sales for residential use.

3.2.2. Ports and economic activities

Historically, human settlements were inferred to abundant and potable water resources in order to palliate technological paucity. Handworks labor and population should be supplied, resort to highly polluting techniques involved (tannery, slaughterhouses, clothiers, etc.) and wastewater treatment had not yet been invented (Leguay, 1999).

Developing landlocked cities were consequently located near large rivers, but this indispensable water could represent a major threat: even the early civilizations soon learned to protect themselves from floods. Upstream dams and channelling were beyond their ability for large streams but were rapidly set up for minor rivers.

Moreover, hydrologic droughts, historically mainly due to lack of precipitation, had dramatic consequences: even if the water supply was the main problem, maintaining a navigable depth was progressively more difficult when the size of boats increased: particularly during the 19th century, large cities accommodated their ports with low dams in order to allow barge circulation and dug artificial coves for barge mooring. Combined with the industrial era's perception of shoreline development (i.e., clear-cut logging of riparian trees), numerous cities interconnected them to an anthropized fluvial network whose shoreline erosion accounted for about half of the sediment load of urban streams (Trimble, 1997), which accumulated upstream of the urban dam.

For coastal cities, sedimentation could be due to urban activities and, as for dams, to lentic areas bought for naval security reasons. Old-named "heavens", such places could be connected to estuaries but were more often built on the shore for long-term mooring and in order to provide a calm harbor. Such conditions enhanced suspended matter deposition. Moreover, sediment could receive water or wastewater from the shore. However, boating and other naval activities induce additional pesticide consumption: antifouling is mainly performed by using pesticides against algae and shellfish. Numerous publications provide information on past and modern pesticide use, from tributyltin and its derivate to current mixes. For example, Okamura *et al*. (2003) mentioned Irgarol use in Japanese ports, where the highest Irgarol concentrations are observed. Carbery *et al*. (2006) noted the same pattern in the Caribbean harbors of the Virgin

Islands and highlighted amateur mixes made with Irgarol and diuron. Their sampling included sediment, where the maximal concentrations were obviously found.

Whether river port or sea port, sediment accumulation has been observed, and sediment is very well known for accumulating metallic (Cooper & Harris, 1974) and organic (Karickhoff *et al*., 1979) contaminants: sedimentation due to human activities induces contaminant storage in populated areas (Devault *et al*., 2007), where pesticides are only one of several contaminants. Because of the urban context, such sediment could reach high biocide concentrations leading to contamination hotspots and, for river ports, contaminating the aquatic environment downstream during major floods.

Chauvel (2006) asked industrialists, including the transport junction, about their pesticide consumption. In descending order, industrialists consider pest control to be useful to:

- limit fire risks (herbicides against brambles and thickets), completed by the third item in this list.
- close behind fire risk, esthetic considerations are brought up: weeds are a sign of disorder, decrepitude, inactivity and, finally, abandonment. On the contrary, business and work areas have to impress competitors, customers, suppliers and employees with an image of organization, hygiene, and activity.
- Weed development could be an obstacle for rescue operations. A practical argument could be based on risks from animals on legacy obligation or on inner safety committee requirement.
- Equipment and structure alteration. Depending on the equipment and structures involved, esthetic concerns could predominate. The risk from animals is the main risk: electrical installations (power plants, airports, etc.) are sensitive to damage by animals.
- Risk of pest invasion. Some of the industrialists surveyed were in the food processing industry, but this could be redundant with the previous item.
- Health risk. Only 6% consider this risk as sufficiently pertinent to justify pesticide use (Chauvel, 2006).

4. Aspersion of pesticides and the consequences of pesticide transfer

In the urban context, use of aspersion depends on the substratum. Agricultural practices could be adapted to lawns and parks. To a lesser extent, clay sidewalks and paths could be treated with the same equipment. However, considering impervious substratum such as asphalt, pavement, concrete slabs or roofs (Van de Voorde, 2012), using the same techniques is not viable: urban pesticide spraying occurs in "tiger stripes" on impervious surfaces, which does not facilitate comparison with agricultural uses.

The example of railways should be cited: high-speed trains, whatever their model (the TGV in France, the Shinkansen in Japan, the ICE3 in Germany) could be struck by weeds growing on embankments and because of the high speeds attained by these trains, this could damage the rolling stock. Consequently, railway companies are identified as potentially significant polluters.

To avoid aquaplaning, rainwater should be rapidly evacuated. Roads are therefore directly connected to sewers. Even if safety imperatives prevail, this direct surface runoff could generate serious consequences (see below).

In France, 190 airports, covering between 50 and 2000 ha (Chauvel, 2006) have paved ground totaling more than 50% of nonagricultural use. Approximately 1 million km of highways and freeways cover France, combined with all types of roads covering approximately 713,500 ha, including 145,000 ha of grassed surfaces (Chauvel, 2006), about 6% of France's total surface area.

Considering railways, information is still heterogeneous except for systematic control embankments: Schweinsberg *et al*. (1999) estimated pesticide input at approximately 8–10 t/ha, but the French railway company only declares 3 kg/ha (Blanchoud *et al*., 2004). This result highlights how linear to surface expression could bias reasoning: in France, cumulated railways are about 85,000 km long (Chauvel, 2006).

Indeed, considering maintenance of impervious surfaces, users try to control weeds growing in fissures or interfaces between impervious surfaces. This type of application also depends on fissure/interface location: along a wall, weeds could be considered as less anaesthetic or impeding than along a gutter (Zadjian *et al*., 2004); grassed suburban sidewalks are regarded with more tolerance than city center sidewalks. Narrow cracks in the substratum are sprayed, targeting weeds, including the impervious surroundings, a practice that is more widespread than in the agricultural context, considering weed biomass as well as surfaces: a survey of the Californian Department of Pesticide Regulation (Fossen, 2009) noted that 60% of pesticide use in urban areas occurred on impervious surfaces.

4.1. Runoff transfer

Blanchoud *et al*. (2004) estimated pesticide runoff from agricultural areas to be between 0.1% and 2.4% depending on runoff conditions. Concomitantly, under the same rain conditions, runoff in urban areas was between 0.8 and 6.7%. These results are confirmed by Wittmer *et al*. (2011a) who observed that rural pesticide runoff was between 0.4% and 0.9% when pesticide runoff in urban contexts was about 0.6–15%. The transfer rate in agricultural contexts is in agreement with Clark & Gloosby's review (2000), who estimated agricultural exportation between 1 and 4%, Leonard (1990), who estimated agricultural runoff at about 2%, and Bro-Rassmussen (1996), who determined maximum runoff in field conditions from about 0.5 to 5%. It also integrates pesticide losses from plots where storm events occurred such as highlighted by Louchart *et al*. (2001) and Revitt *et al*., 2002. Thus, Wittmer *et al*. (2011a) propose that pesticide runoff from urban areas could be considered as one order of magnitude greater than in agricultural areas. This estimation seems to be in accordance with the literature. The agricultural maximum transfer rate observed in blind conditions (for diuron) is close to the urban maximum transfer rate, to our knowledge, at the watershed scale (45.1%, Revitt *et al*.,

2002) but is clearly much rarer than in the urban context. The 6% transfer runoff integrating agricultural and urban areas of a whole watershed proposed by Blanchoud (2001) seems to be consistent.

Apart from runoff, pesticides spread on limited-adsorption surfaces will be exposed to other processes. However, to our knowledge, no study has specifically detailed the abiotic fate of pesticides in these conditions. Indoor conditions will be detailed in another chapter.

4.2. Lixiviation transfer

Less studied and less obvious, the impact of urban areas on lixiviation remains significant (1) because the lixiviation volume is minimized and (2) because pesticide transfer to groundwater differs comparatively to other land uses (Trauth & Xanthopoulos, 1997).

As previously indicated, water cycles in urban areas are modified: the contribution to groundwater is halved compared to the natural water cycle. Compared to urban pesticide use, groundwater could be more contaminated than under agricultural land. Thus, it is possible to identify the urban impact on groundwater just as it is possible to identify the urban impact on surface waters.

Bruce *et al*. (1996) distinguished residential, commercial, and industrial areas in the urban impact on groundwater. Commercial areas have a greater impact on groundwater because of ornamental plants as well as roads and parking lots, while residential areas are more marked by the needs of ornamental plants and industrial areas by impervious surfaces. Residential areas showed higher contamination levels than industrial areas. However, Trauth & Xanthopoulos examined this segregation: urban areas mix industrial plants (i.e., point source contamination), roads and sewers (linear contamination), and allotment areas (surface contamination): groundwater contamination is not the faithful reflect of the surface one. Nevertheless, statistical results on studies on wells have shown that pesticide concentrations were higher in urban areas than in rural areas. Even if some pesticides are found more frequently in urban areas, statistical consistence is impacted by the number of wells. Malaguerra *et al*. (2012) outline groundwater contamination via the groundwater table and sediment from contaminated streams caused by enhanced runoff in urban areas. Inversely, because of less vertical water transfer due to impervious surfaces, leaching could be slowed, favoring degradation and lateral water transfer, mixing groundwater contaminants (Malaguerra *et al*., 2012).

5. Resident exposure to pesticides spread in urban areas

Pesticide use in urban areas is a major concern for the aquatic environment as well as for human health (Van Maele-Fabry *et al*., 2011). The influence of water contamination resulting from urban pesticide runoff is greater on an aquatic environment than on human health, and food contamination is due to agricultural applications of pesticides. Consequently, the main exposure of urban residents by pesticide spread in urban areas stems from air contamination

(Ragas *et al*., 2011). Moreover, except for pesticide use in urban areas compared to agricultural areas, the urban context favors human contamination by atmospheric pesticides. Due to hydrophobic patterns of the majority of pesticides, contamination by dust is the main source of contamination by air. The aim of the present chapter is not to propose a review of the abundant literature on contamination by pesticides and associated dusts. Appropriate reviews exist, e.g., Schneider *et al*. (2003), Bradman & Whyatt (2005), Garcia-Jares (2009) Kanazawa & Kishi (2009) and Karr (2012). The aim is rather to explain why the urban context encourages human contamination.

In short, buildings are enclosed, windless, sunless, partly septic spaces where dusts can be trapped and accumulate, particularly in fabrics such as carpets (Obendorf *et al*., 2006): 80% of pesticides found indoors are found in clothes, particularly shoes (Quiros-Alcala *et al*., 2011). Moreover, degradation occurs less indoors than outdoors (Roberts *et al*., 2009). Other variables independently associated with dust levels included temperature and rainfall, storing pesticide products in the house, housing density, imperfect housecleaning, and air conditioning (Harnly *et al*., 2009). Farmworkers expose their families more than other professional categories (Quiros-Alacala *et al*., 2011); consequently, in suburbs, municipal service employees and private gardeners could be considered as possible vectors to their relatives. Weschler & Nazaroff (2008) outlined the relationships between gaseous organic chemicals, including pesticides, and dust contamination: solubility and Koa (partition coefficient between octanol and air for chemicals) successfully describe gaseous pesticide contamination, in contrast to other molecules (Schoeib *et al*., 2005). Clothes abrasion and other contaminations (paint coating) occur indoors, promoting indoor pesticide content and some of the organic matter in dust, such as cotton linters, may differ substantially from octanol in terms of sorption of gas-phase Semi-Volatil Organic Compounds (Weschler & Nazaroff, 2008). Direct contact of dust with polluted surfaces seems to be enough to pollute dust (Clausen *et al*., 2004). Moreover, high indoor temperatures induce chemical volatilization, and the difference between the laboratory temperature for Koa determination and the private indoor temperature could be significant. Passive air sampling does not efficiently inform about long-term contamination because of passive samplers (Weschler & Nazaroff, 2008) and quantification thresholds.

Blanchoud (2001) estimated agricultural pesticide amounts used on the Marne watershed at about 5200 t/year, urban use at about 62.5 t/year and atmospheric amounts at about 0.5 t/year. But global contamination should not be ignored: MCE (2003) estimated that Rhine valley inhabitants, by breathing, were twice as contaminated by pesticides than if they drank 1.5 L of water with close to 0.5 µg/L total pesticide concentration, i.e., the maximum allowed concentration by surface water quality norms. Moreover, gaseous pesticides are directly bioavailable compared to pesticides associated with particles, which sequester more than 99% of the main pesticides (Koc and Kow>2). Studies on pesticide exposure mainly target farmers and pesticides used in agricultural areas (Mercadante *et al*., 2012). Considering the issues at hand, data on public exposure to urban pesticide use are rare, even if studies are currently in progress.

Population exposure to contaminated particles or volatile pesticides is more than ever an issue because this exposure occurs as much at home as at work, and because enclosed living spaces affect every age group.

Air contamination data is still too rare and incomplete, and would benefit from further study.

To pesticides designed to protect crops, one must add a large number of biocides designed for heath or esthetic uses (household products, paints containing algaecides, etc.): the nature of these pesticides is not well understood by users. Because the sense of sight prevails over the other senses, the most readily perceived pollution is air pollution, associated with transport. Coupe *et al*. (2000) assert that high oxidative conditions in urban areas compared to rural areas (Finlayson-Pitts & Pitts, 1986) promote pesticide oxidative degradation. There is no evidence of a significant urban influence.

6. Progressive pesticide awareness of urban pesticide use

For Denmark and the Netherlands, the first monitoring programs demonstrated evidence of water contamination. Depending on its groundwater for drinking water, in 1995 Denmark discovered its groundwater pollution level. For the Netherlands, water pollution was striking because of the Meuse River contamination, which resulted in a ban, forbidding water intake for 7 weeks (1993 and 1994), while this country depends on surface water for 40% of its drinking water, soon to rise to 50%. In wooded Sweden, the threat to human health was the main driver because Swedish forestry and roadway services air-applied Agent Orange, a 2,4-D and 2,4,5-T formulation known for its mutagenic potential. Concerned by Agent Orange use and air-spraying, the population continued to debate about the daily place of pesticides after Agent Orange's definitive ban (1977). The first monitoring campaigns were carried out in 1985 and revealed water resource contamination, leading to early and radical directives (Ulén *et al*., 2002).

Pesticide awareness differed in the largest countries. For agricultural countries such as the United States and France, pesticide awareness came early but was mainly associated with agricultural use. In Germany, the negative effects of pesticides were avoided by early plant protection legislation: the first legislative provision was decreed in Germany in 1919 and was implemented in 1968. Thus, weed control by herbicides is forbidden on hard surfaces without local authorization and only if there is no runoff risk. In this case, plant protection control programs determine the few available chemicals. For the US and France, pesticide contamination evidence dates back to the 1960s. Associated with agriculture, pesticide use was rarely reported in urban areas until the 1990s when extensive monitoring, directives in other countries, and early scientific publications (Cole *et al*., 1984) awakened awareness. The Nationwide Urban Runoff Program, prepared between 1978 and 1980, carried out between 1980 and 1982, provided the first public information on urban contamination in the US. In comparison, the first French publication on urban pesticide contamination is Chevreuil *et al*. (1996), and was still focused on agricultural contamination; even if the Water Law was decreed in 1992, compatible urbanization was taken into account in 2004 (Diren, 2010). The European

Union is a key factor in French pesticide awareness. Like the above-mentioned countries, the UK did not experience a catalyst event leading to massive pesticide awareness. Without previous legislation as in Germany, and without agriculture importance like in the US and France, urban pesticide use would have been more evident given Greater London's importance in UK land use. However, British awareness seems limited and related publications are scarce (only Rule *et al*., 2006, and Stuart *et al*., 2012).

The virtuous pesticide approach is performed in agreement with the European Union: at the same time, legislators follow European directives such as the Council Directive 91/414/EEC (EEC 1991) and the Water Framework Directive (EEC, 2000), and support forums on amenities or pesticide representations. However, this process is more efficient in countries with leading governance such as France: British self-regulation practices and the tradition of voluntarism make them less easy to apply (Grundy, 2007).

Initiatives could be combined as is done in the US. First of all, professionals are involved, then private applicators. Consequently, since 1993, the US has established a 2-year license for spraying pesticides depending on member states' initiatives, presenting different process levels. For example, Idaho, Georgia, and Minnesota have established a voluntary program for the publication of a pesticide sale and use database. Idaho and Georgia follow Urban Pest Management Programs in order to involve individual applicators.

Although Germany's 1919 plant protection decree was a notable base for environment protection, the country continues to strengthen its pesticide reduction policy even if detailed data for urban herbicide do not exist. Parenthetically, annual consumption of pesticides used in part in urban areas is about 230 t. Kristoffersen *et al*. (2008) estimated glyphosate, the main pesticide used in urban areas, at about 2 t/year. Finland's use is estimated at about 5–6 metric tons of active molecules per year (*Ibid.*); the substances allowed are limited and use of very toxic pesticides is limited to qualified persons.

Pesticides used in urban areas are limited; for example, diuron is often forbidden in Europe Union countries and the Canadian province of Ontario (decree 63/09, 4 March 2009, enforced 22 April 2009) banned all pesticides use for esthetic purposes, but some limited uses, such as on golf courses, are allowed. Golf courses require intense pest control and artificialized surfaces, with pesticide transfer close to urban areas. In France, greens cover about 20,000 ha. Considering the 550 golf courses in France, the average surface of a green is about 36.4 ha. Even if economic arguments, ecological concerns, and society's growing awareness are influencing golf course managers taking these concerns into account, the results from such sites should be considered with caution, due to divergent goals or the risk of different interpretations.

The ultimate level of urban pesticide use awareness is differential taxation and alternative innovation. The Netherlands and Denmark are the most forward-looking countries for alternative development, followed by Sweden, the leading European countries for environmental issues.

In addition to legislation, the importance green political parties or related are a better reflection of the population's awareness of ecological concerns, as expressed in elections in the number of deputies for a given population: Germany sent 22 ecologist deputies to the European Parliament, France 19, Sweden, the Netherlands and the United Kingdom 5, but, in contrast to others, the United Kingdom's deputies are mainly autonomists. With these results, the political interest in the environment could be considered as moderate (Kristoffersen *et al.*, 2008). Thus, adoption of an ethical attitude will be limited until citizen support is expected. For example, despite its legislation strictly controlling urban pesticide use since 1954, Finnish people do not show a willingness to complete its legislation by greater amenity pesticide control (Kristoffersen *et al.*, 2008). Moreover, hard surfaces or the status of amenity areas could curb initiatives: in the United Kingdom, administrative land fragmentation results in local authorities being responsible for weed control (Grundy, 2007).

7. Technologic alternative

Road shoulders were mowed in certain places up to the 1950s, although hay production was declining at this time. Green shoulders limit soil erosion and consequently prevent road sap, helps drivers see curves and anticipate the course of the road, allows a good visibility of signs, protect from wind, and prevent monotony for drivers and eyesores for residents. However, walkers, wildfowl and rain require road shoulders to be flush cut. Margoum (2003) highlighted how ditches could enhance pollutant retention. Considering pesticide costs and low user solicitation, highway companies could notably reduce their biocide budget using alternatives to pesticides (at least 50%, Mahe (2007)).

Based on Table 1 (Marque & Chabaud, 2006), in order to control at least 8- to 10-cm-high weeds, mowing seems to be the best alternative. Mowing does not induce soil or root lifting, and cutting at an appropriate height could avoid passage: flush cutting weeds too short could harm low-growth perennial plants, which inhibit high-growth annuals such as allergen ambrosia. Moreover, perennial weeds are often more endemic than annual weeds, contributing to biodiversity promotion. Mowing seems to correspond to private and public professionals' financial means and satisfaction surveys highlight its popularity (Mahe, 2007).

In Germany, a system has been developed, the Rotofix, a hand-operated roller sprayer as an alternative to spray a zone for a single plant. Appropriately used, it could reduce herbicide volume by 75–95% (Hermanns *et al.*, 2006).

Instead of using pesticides, public authorities could employ other molecules, such as acetic, citric and pelargonic acids on hard surfaces. Although it is used in Germany, acetic acid is also prohibited in 50% of Swedish municipalities and is only allowed in the Netherlands when there is no runoff risk.

Ground cover could be an alternative (Table 1), if the ground use is amenable (Marque & Chabaud, 2006). Mulching and covering with plastic drastically limit weed growth, but could be too aleatory (vegetal wood and cloth covers), temporary (vegetal and cloth), fire-prone (vegetal, polypropylene and cloth), unaesthetic (polypropylene, vegetal cover with time), difficult to deploy (minerals are heavy, vegetal covers need time), and require maintenance (vegetal and mineral covers).

Table 1. Qualitative and economic analysis of alternative preventive methods (from Marque and Chabeaud, 2006).

Technique	Material	Advantages	Disadvantages	Raw material cost	Productivity cost	Number of input (/year)	Cost/m²	Durability
Mulching	Vegetal matter	Ease of use Natural visual Water protection Matter organic input	Lengthy deployment; Volatile; Animal deterioration; Annual complement; Visual with time; Risk hazard; Dubious herbicide	0.2 to 2€/m²	3€/m²	0.1*	3.3 to 6.2€/m²	1 to 5 years
Mulching	Mineral matter	Heavy products	Ease of use but lengthy; Water protection; Animal deterioration; Needs complement; Herbicide	2.5 to 3€/m²		1*	5.5 to 6€/m²	5 years et +
Mulching	Plastic matter, polypropylene	Easy and fast to deploy; Water protection	Impervious; Visual controversy; Film eviction problems Risk hazard; Herbicide	0.25 to 0.70€/m²	0.70€/m² for plantation + for eviction	0.25 to 0.15*	0.95 to 1.40€/m²	5 to 7 years
Mulching	Ligneous slabs	Ease of use	Impervious; Dubious herbicide	0.5 to 2.5€ each	0.60€ to 1.10€ / each	1	1.10€ to 3.60€/m²	2 to 4 years
Mulching	Plant fibers and rags	Ease of use; Water protection; Recycling	Fragility products; Visual; Risk hazard; Dubious herbicide	1 to 4€/m²	1€/m²	0*	2 to 5€/m²	2 to 5 years
Ground cover	Plants	Natural; Esthetic; Good cover with time	Needs protection the first years; Maintenance	10m€/m²	3.5€/m²	Retouches*	13.50/m²	10+ years
Chemical	Anti-germinatives	Easy for granule; Fast use; Efficient;	Pesticide use; Follow good practices and framework legislation;	0.05€/m²		1	0.11€/m²	1 year
	Foliate	Selective products	Qualified staff	0.01€/m²	0.06€/m²	1 to 3	0.07€ to 0.21€/m²	Following regrowth

7.1. Mechanical alternatives

Brushing can be used only on impervious and clean surfaces. At best, the rotation of the bristles extracts part of the roots. However, coated surfaces are abraded: asphalt near fissures could be snatched, deteriorating bristles. Pavements should be cohesive and regular but slipping could occur when wet. Moreover, steel brush tests have demonstrated the level of noise and vibration is incompatible with good working conditions and urban use (Hansson *et al*., 1992 in Rask & Kristoffersen, 2007). Brushes are only made in polypropylene. Despite brushing's efficiency, Lefevre *et al*. (2001) and Wood (2004) do not recommend it for long-term use but Lefevre *et al*. (2001) and Hein (1990) propose to use it for heavily weeded areas.

Rotative clogs comprise a heavy metal cylinder rolling on the ground and extracting roots. They are only used on pervious surfaces, which should be tamped after the application, an expensive step. The surface is severely abraded: rotative clogs could be limited to clay surfaces (Hamelet, 2004).

Sweeping, whether or not it is mechanized, even in gutters, could be useful, despite the number of sweepers required, and is a no n-hermal alternative (Hein, 1990; Parker & Huntington, 2002; Hansen, 2004): the advantages of cleaning could justify the price of optional engines or numerous teams. Lefevre *et al*. (2001) considered that seven to ten operations per year were very efficient for controlling weeds in temperate climates.

Harrowing is still efficient on gravel surfaces: easy to use, inexpensive to purchase, maintain, and deploy, in 1992 it led to banning herbicides for churchyard treatment in Denmark (Tveedt *et al*., 2002, in Rask & Kristoffersen, 2007).

Paradoxically, human mechanical work, whether it is used marginally or institutionally, seems to keep up for limited surfaces (Angoujard *et al*., 1999): the Versailles municipality organizes hoeing teams of seasonal workers (Mahe, 2007). The main obstacle is the cost of labor for developed countries with high labor costs, but this obstacle could be reduced in emerging or developing countries where sweeping appears to be a reliable alternative to herbicide use. However, such practices could be considered as retrograde and even degrading.

7.2. Thermal alternatives

Thermal alternatives use heat to scorch or burn off weeds. Heat could be obtained with sun (solarization), high-pressure steam, sugar foam, infra-red, freezing or gas flames (Table 2).

Globally, thermal uses require many passages (Rask, 2012) and are highly energy-consuming. Treatments are more effective on low-growth weeds and roots are scarcely damaged. The driving speed must be slow for an effective treatment. The main target of thermal alternatives is to expose pesticides to warm conditions, so as to degrade them. However, especially when the vector of the fluid, i.e., steam, warm water or a warm mix, the temperature reached should not be high enough to degrade or even mineralize the pesticide, but could enhance volatilization. This phenomenon is known to significantly affect the fate of some pesticide families. In the urban context, due to a lesser adsorption phenomenon leading to enhanced pesticide runoff, ground temperature, H Henry's constant greater than 10^{-5}, and the effects of Raoult's

Technics	Material	Advantages	Disadvantages	Investment in material	Productivity	Number of passages	Annual cost	Consumable
	Hoe	Ease of use; All weather; Handy; Very low investment	Number of passage; Labor-intensive; Limited to small surfaces.	20 to 30€	50m²/h	5 to 6	0.40€/m²	
	Rotative clog	Low investment; Ease of use	Number of passages; Only on pervious surfaces, even degradation					Gasoil
	Rotative brush	Moderate investment; Ease of use; Effective on pavement	Substratum and joint degradation; Number of passages	4k to 5k€	1000m²/h	4 to 6	0.36€/m²	Gasoil
Mecha-nical	Sweeper	Cleaning, Sharable cost	Average to very high investment; Number of passages; Joint degradation; Limited to gutters	2.65k to 90k€	2600ml/h	8 to 12	0.12 €/m²	Gasoil
	Mowing, small equipment	Moderate investment; Ease of use; All weather; Handy	Number of passages; Labor-intensive; Not for large surfaces; Fossil-energy dependent	200 to 600€	450 m²/h	3 to 4+	0.23 €/m²	Two-stroke gasoline
	Mowing/ mulcher, big equipment	All weather; Handy; Effective on ligneous	Average to expensive investment; Consumption of fossil energy; Visual on ligneous	2 to 20k€	3 to 6000 ml/h	1 to 2	0.12 €/m²	Gasoil
	Infrared	Medium investment; Ease of use; Handy; Useful on pervious as impervious; Manual or mechanized	Number of passages; Risk hazard; Only for seedlings and monocotyledons; Consumption of fossil energy; Greenhouse gases	3.3k to 9.5k€	2 km/h 1000m²/h	6 to 8	0.24 €/m²	Propane (1kg of gas/h/burner).
	Flame	Low to moderate investment; Ease of use; Manual or mechanized	Number of passage; Risk hazard; Consumption of fossil energy; Greenhouse gas generation	0.5 to 6.2€	3 to 5 km/h	4 to 6	0.22 €/m²	propane 2kg of gas/h/burner
	Steam	Efficient on impervious surface; Handy; Mechanized; Polyvalent; Cleaning effect	Expensive investment and water; Moderate efficacy on settled weeds; Number of passages	17.5k to 44k€	1600 ml / h 1100 m² / h	3 to 6	0.25 €/m²	Eau (4 to 500 l /h) gasoil
Thermal		Disinfection	Diesel fuel consumption; Ground speed 0,7 to 1 km/h					Gasoil (4 to 7L/h)
	Warm water	All weather; Handy; Polyvalent; Hose Effective on bryophytes	Average investment; Number of passages; Only on leaves; Water and diesel fuel consumption; Ground speed 1 to 2 km/h	15k to 18k€	1000 m²/h	4 to 6	0.25€/m²	Water (3 to 400 L/h) gasoil (4 to 5L/h)
	Warm foam	Useful on pervious and impervious surfaces; Handy; All weather; Number of passages	For rent only; Mainly effective on leaves; Water consumption; Diesel fuel consumption; Ground speed, 3 to 5 km/h	0.7k€/d*; 0.9k€/w; 3k€/m; 13k€/6m; 21k€/y;48k€/4y	350 m²/h	3 to 4	1€/m²	Water (15 to 50m³/ha); Gasoil (5 to 6L/h); Foam 100€/d; 0,2 to 0,4% additive
	Foliate	Low to moderate investment; Ease of use; Pervious and impervious surfacees; Manual or mechanized; Dose modulation following the weeds	Weather dependent; Run-off risk on impervious surfaces; Number of passages; Average efficacy on certain plants	50 to 5k€	1000 m² to 2000 m²/h	3 to 5	0.11 to 0.17 €/m²	Products 35€ Water Gasoil ***
Chemical	Foliate + anti-germinative	Low to moderate investment; Number of passaes; Manual or mechanized; Curative and preventive	Weather dependent; Pervious surfaces only	50 to 5k€	1000 m² to 2000 m²/h	1 + 1 retouch	0.12 to 0.18 €/m²	Products 400€ Water Gasoil ***
	Foliate+ residuary			50 to 5k€	1000 to 2000 m²/h	1 + 1 retouch	0.15€/m²	Products 150€ Water; Gasoil ***

Table 2. Qualitative and economic analysis of alternative curative methods (from Marque and Chabeaud, 2006). *Short-term rental including equipment+driver/technician **Equipment Rentals vehicle or technician without applicator ***If mechanized implementation.

law, pesticides could be exposed to enhanced volatilization (Burkhardt & Guth, 1981). For Scheyer *et al.* (2007ab) and Delaunay *et al.* (2010), high amounts of volatilized urban pesticides are notably observed in urban air but are too limited to induce long-distance contamination or to significantly pollute agricultural areas when farmland pesticides are found on the same order of magnitude in agricultural areas as in urban areas.

Considering *solarization*, two limits are identified. First of all, the weather should be sunny (at least 250 h/month) and shade should be avoided (due to other weeds). Also, a large amount of plastic waste is generated (Cheroux & Serail, 2006).

Due to the nature of impervious surfaces, i.e. mainly dark asphalt, solarization could lead to extreme temperatures (asphalt fusion temperature: between 90°C and 110°C). Many pesticides, particularly herbicides, could lyse at such temperatures, but no study has investigated this question. Even if the sunshine does not induce high temperatures, photolysis could occur but no direct evidence has been found in the literature for this special case. However, the long-term experiments conducted by Jorgenson & Young (2010), Jiang *et al*. (2011) and Jiang & Gan (2012) do not mention photolysis of urban pesticide, but experiments examined low photolysis-sensitive pyrethroids. However, the observed loss is far from being as fast as expected with less than 1 h DT50 photolysis at neutral pH (Fossen, 2006).

High-pressure steam application requires substantial quantities of water and a substantial financial investment. Its efficacy is poor (Daar, 1994) because of perennials. Foam could be used instead of water, made of coconut sugar and corn sugar, to enhance warming duration and subsequent efficacy (Daar, 1994). Numerous applications are required.

Gas flames alternative use has the advantage of being an intuitive and light (Rask, 2012). However, this alternative is expensive (substantial gas consumption) and may even be a source of fire danger (Wood, 2004). It is the most commonly applied thermal weed control method on hard surfaces. In Germany, a train equipped with flame weeders has been elaborated for railway embankments (Kreeb & Warnke, 1994).

Infrared radiation is the most effective non-chemical control method and economically comparable to herbicide treatment (Augustin, 2003, cited by Rask & Kristoffersen, 2007), but radiators are very expensive, brittle, and inoperative for dense vegetation (Ascard, 1998).

Other alternatives have been tried: laser (from Couch & Gangstad, 1974, to Heisel *et al*., 2002), gamma radiation (radioactive), UV radiation (nullified by mutagenic and fire hazards), microwaves (hazardous and need 1000–3400 kg diesel/ha for a significant effect according to Sartorato *et al*., 2006), electrocution (fire hazard for the surrounding terrain, electrocution risk for operators and passers-by, and a high amount of electricity needed, but this could be an alternative for railway pesticide uses). None of these methods currently presents consistent results.

8. Conclusion

The use of pesticides is still too directly associated with agriculture, a clear cultural barrier for those countries that are built on a strong dichotomy between the countryside and the city, the latter needing to be maintained. However, in developed countries, urban sprawl seems to be the main driver of water resource contamination. Outer urban areas are the most vulnerable to pesticide use: they do not have a water collection and treatment system as developed as those in city centers, while suffering from a greater level of pesticide pressure than is found in agricultural areas. However, studies on pesticide representation are mainly done in agricultural areas, and their urban equivalents are rarer.

Populations believe that the impact of indiscernible actors of pollution does not exist. Moreover, the fate of pesticides in urban substrates (asphalt, etc.) is not sufficiently known: recent studies on contaminated concrete and paint have shown gaps in our understanding of sorption, volatilization, and photolysis processes.

The descriptors of the pesticide pressure are lacking but also in need of improvement: comparing the surfaces covered by agricultural machinery spraying to urban “leopard spot” or “tiger stripe” spreading is not relevant. This lack of standardization can be found in studies seeking to highlight the performance of alternative techniques for spreading, which nevertheless seem capable of improvement.

Nonetheless, new curative or preventive tools could provide effective alternatives to pesticide use. The pivotal quality of alternative strategies lies in the choice of matching the tool to the substratum. However, this pas de deux is essential for limiting pesticide contamination.

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