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Residues of diazinon in Ab-bandans supplied by Babolroud, Talar and Siaroud Rivers, Iran

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In this http://www.wessex.ac.uk/components/com_forme/uploads/ study, diazinon, as an organophosphorus pesticide, was measured in nine Abbandans (man-made wetland) in Mazandaran Province: Kharajisha, Ramenet, Esmaeelkola, Kordkola, Shrag-e-Larim, Anarmarz, Roshandan, Galeshkola and Langoor from April 2010 to March 2011. A total of 216 samples were taken from nine abbandans of the South Caspian lowland. Samples analyzed revealed that diazinon is observed frequently in aquatic ecosystems throughout the year in summer, mostly. All abbandans except Kharajisha have shown similar trend annually. Kharajisha as an aquatic system is fed by Babolroud River, which is exposed nearly three times to diazinon concentration than others in summer. In other seasons, it decreases gradually so that winter and spring have lower amount. Due to the vast expansion of agricultural fields in the study area, various contaminants such as diazinon are leached by rainfall, irrigation and drainage activities and finally are conducted to the adjacent rivers and abbandans. These findings indicate that diazinon is widespread in the environment and can possibly have adverse effects on aquatic ecosystem health. Thus, development of irrigation and drainage efficiency and some environmental observations under the small land holding condition could diminish the negative impacts and consequently serve the ecosystem balance in this region.

Key words: Diazinon, Babolroud, Talar, Siaroud, Ab-bandan.

INTRODUCTION

One of the most important types of wetland in the South Caspian lowlands is the “Abbandan”, a small, man-made reservoir or flooded rice paddy with a luxuriant growth of underwater vegetation. These shallow wetlands, that vary in size from 3 ha to 1,000 ha and with mean depth of 1m, provide excellent feeding and roosting areas for large numbers of migratory waterfowl. Most were originally built as temporary water storage areas to provide water for irrigation during the dry summer months. They also provide habitat for edible fish such as silver carp (*Hypophthalmichthys molitrix*) and grass carp (*Ctenopharyngodon idella*). A total of 506 abbandans were estimated in Mazandarn Province, having 14811 ha area. They are distributed through 635 villages and so benefit about 50000 families. The study area, located in the Southern Coast of the Caspian Sea along the Mazandaran Province of Northern Iran, is one of the most productive agricultural regions in the Middle East. Within the Mazandaran Province, a large quantity of pesticides is used to protect crops from pests; in 2006, almost 30,000 tons of pesticides were used in the agricultural areas (Ahmadi et al., 2011). Most areas of plain regions in Mazandaran Province are planted with rice, and different pesticides and fertilizers are used at high

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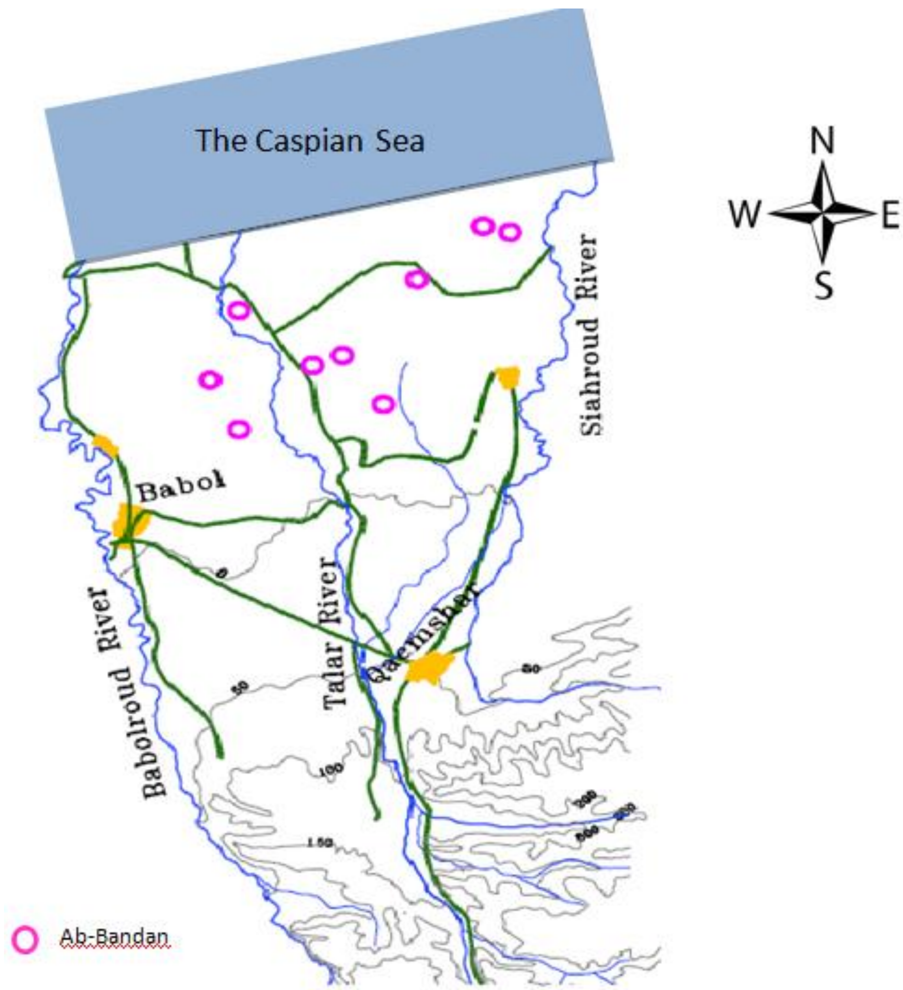

Figure 1. Map of studied area and sampling sites.

densities to increase the yield of production. Study area extends from Babolroud to Siaroud. Many abbandans created in this area such as Kharajisha, Ramenet, Esmaeelkola, Kordkola, Shrag-e-Larim, Anarmarz, Roshandan, Galeshkola and Langoor were selected to measure diazinon toxin (Figure 1). All mentioned abbandans feed mainly by Babolroud, Talar and Siaroud rivers and somewhat through rainfall, drainage and underground water.

Diazinon is an organophosphorus (OP) compound and a broad-spectrum insecticide that is used as a pesticide in agriculture and non-agriculture activities. This pollutant has been detected in freshwater, seawater, point-source discharges, and storm-water runoff in urban and agricultural areas (Kawai et al., 1984; Talebi, 1998; Bailey et al., 2000; Konstantinoun et al., 2005; USEPA, 2005; Luo et al., 2008). Diazinon is the main insecticide used to control stem boring caterpillar of rice (*Chilo supressalis*), lice, blowflies, ked, ticks in sheep, cattle, goats, dogs and so on. It controls aphids, caterpillars, moths, butterflies, various worms, locusts, grasshoppers and scale in pastures, orchards, vegetables and field crops. This finding indicates that pesticides, especially diazinon, are widespread in the Northern provinces and can possibly have adverse effects on aquatic ecosystem health.

It is moderately persistent but also mobile in the environment. After December 31, 2004, it became unlawful to sell outdoor, non-agricultural diazinon products in the United States (USEPA, 2005).

In the present study, the general objectives are to evaluate seasonal distribution of diazinon in different aquatic ecosystems, identify higher local concentrations to improve suitable agricultural activities toward sustainable development.

MATERIALS AND METHODS

Nine Abbandans including Kharajisha, Ramenet, Esmaeelkola, Kordkola, Shrag-e-Larim, Anarmarz, Roshandan, Galeshkola and Langoor were selected among many Abbandans for measuring diazinon concentration as one of the organophosphorus toxins in Mazandaran. All Abbandans were chosen near rivers in order to establish an equal condition in sampling. All samples were collected

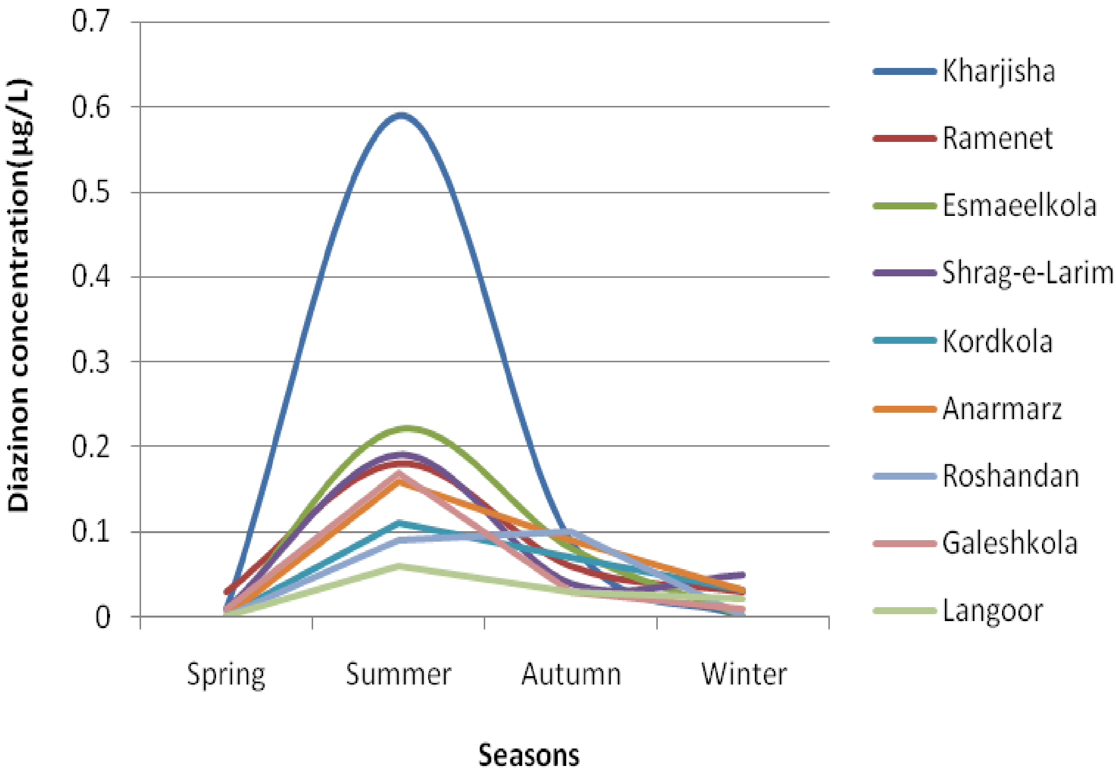

Figure 2. Diazinon seasonal fluctuation in Ab-bandans.

from designated stations every mid-month and collected from middle, across and depth of the Abbandans. Details on the sample sites selection and protocols have been employed as stated by Bartram and Balance (1996) and the general locality of nine selected stations is displayed in Figure 1. Sampling, containers, presservation and the transferring of samples were performed according to the methods described by APHA (1999), USEPA (2007) and Zhang (2007). At each station, duplicate water samples were collected in 1,000-mL glassy bottles. Samples were extracted without any filtration according to the method described by Zweig and Devine (1969) and Zweig (1972). Determination of diazinon in water using solid-phase micro-extraction (SPME) with gas chromatography-mass spectrometry detection (GC-MS) was investigated and detected by a flame photometric detector (FPD).

Extraction of water samples was carried out by direct immersion of the PDMS/DVB fiber in the 4mL sample contained in a 5-mL clear glass vial under magnetic stirring for 45 min at 60°C. Sample agitation was done at 1150 rpm by a magnetic stirrer. Then the fiber was removed from the sample solution and immediately inserted into the GC injector for GC-MS analysis. SPME fibers were desorbed in the splitless mode for 5 min at 250°C. GC-MS was performed with a Shimadzu (Shimadzu, Kyoto, Japan) equipped with a split-splitless injector and connected to a quadrupole mass spectrometer. Data handling and system operations were controlled by the GC–MS Solution software. Separation was carried out using a DB-5 MS capillary column (30 m × 0.25 mm, 0.25 µm, containing 5% phenylmethylpolysiloxane).

For the chromatographic determination, helium (99.999%) was used as the carrier gas at a constant flow rate of 1 mL/min. Injector temperature was kept at 250°C in splitless mode (5min), and oven temperature was programmed as follows: initial temperature 100°C (hold 2 min), 20°C/min to 180°C, and 10°C/min to 250°C (hold 2 min). The total SPME-GC–MS analysis time is 60 min.

RESULTS

Due to the extensive application of insecticides in the Caspian coasts of Iran (rice paddies in particular), this investigation was carried out to obtain the necessary data and information on the concentration of diazinon. Agricultural chemicals can contaminate surface water resources by runoff into streams and lakes or by the lateral movement of chemicals through unsaturated or saturated soil media to bodies of surface water (Shirmohammadi and Knisel, 1994). Accordingly, one of the biggest challenges for Abbandans and rivers are chemical pollution because it is one of the most critical threats to the aquatic ecosystem (Wei et al., 2006). The levels of diazinon concentration in Abbandans and rivers are shown in Figures 2 and 3, respectively. The highest concentration was 0.59 µg/L in Kharajsha and 0.23 µg/L in Siaroud during summer when diazinon is usually used as a pesticide in the rice fields (Talebi, 1998; Ghassempour et al., 2002). As a matter of fact, the highest distribution pattern of diazinon residue occurs in July, when diazinon is used in the fields to provide protection from pests before harvesting. Based on the field survey, harvesting occurs in late September, and thereafter, generally, diazinon and other pesticides do not show up in the study area (Tables 1 and 2).

Diazinon concentrations in this area were less than concentrations from some locations in the world (Ahmadi et al., 2011) (Table 3).

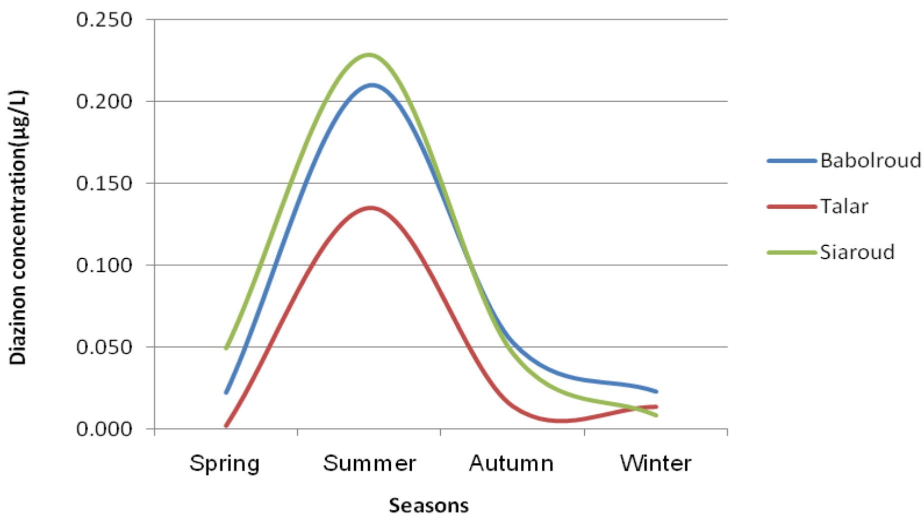

Figure 3. Diazinon seasonal fluctuation in rivers.

Table 1. Mean values of diazinon (µg/L) during four seasons in Abbandans

	Spring	Summer	Autumn	Winter
Kharajisha	0.01	0.59	0.09	ND
Ramenet	0.03	0.18	0.06	0.03
Esmaeelkola	ND	0.22	0.08	ND
Shrag-e-Larim	0.01	0.19	0.04	0.05
Kordkola	ND	0.11	0.07	0.03
Anarmarz	ND	0.16	0.09	0.03
Roshandan	ND	0.09	0.1	ND
Galeshkola	0.01	0.17	0.03	0.01
Langoor	ND	0.06	0.03	0.02

Table 2. Mean values of diazinon (µg/L) during four seasons in rivers.

	Babolroud	Talar	Siaroud
Spring	0.02	ND	0.05
Summer	0.21	0.14	0.23
Autumn	0.05	0.01	0.05
Winter	0.02	0.01	0.01

Table 3. Concentrations of diazinon (ng/L) in rivers from various sites in the world.

Location	Concentration level (ng/L)	Reference
Selangor River, Malaysia	116.1–510.0	Leong et al. (2007)
Kalamas River, Greece	ND to 775	Konstantinou (2006)
Kurose River, Japan	< 2–89	Derbalah et al. (2003)
Susquehanna River, USA	28 (max)	Liu et al. (2002)
Sacramento and San Joaquin River, USA	10–1,690 (max range)	Giddings et al. (2000)
Hendo Khale River, Iran	62–270	Talebi (1998)

more in summer than in other seasons of all nine Abbandans. They have similar trend of Diazinon concentration; it increases in spring gradually and reaches peak in aquatic ecosystems during summer. Similar studies (Khoobdel et al., 2008) have shown significant difference of diazinon and azinphos-methyl concentration during summer and other seasons in Qarahso and Gorganroud Rivers (P<0.05). The results showed that the value of diazinon in water samples was ND to 0.59µg/1. Kharajisha, however, shows more increasing trend as much as 3-10 times compared to others during summer. On the other hand, rivers that feed these Abbandans have also more concentration in summer (Figure 3); in fact, there is direct correlation between diazinon concentration in rivers and Abbandans.

According to British Colombia guideline, diazinon must be lower than 0.1 µg/L to protect fresh water aquatic life from short-term and lethal effects (www.env.gov.bc.ca). To protect fresh water aquatic life from long-term and sub-lethal effects, Diazinon concentrations should be lower than 0.003 µg/L and our results exceeded this value. In the Caspian Sea, there are six commercially valuable sturgeon species, four of which produce 90% of the world's caviar (Hosseini et al., 2008). The Babolroud River is one of the most important breeding habitats for sturgeon species and it provides spawning grounds for them. The high concentration of diazinon pesticide is a threat to the Babolroud River ecosystem and may contaminate the fish products consumed by humans.

Diazinon application decreases when cultivation season ends and its concentration decreases concurrently in aquatic ecosystems. As a matter of fact, rivers are not the only source of Abbandan's Diazinon, so agriculture drainage of adjacent lands also enters into them. For example, Siaroud has more concentration in summer; so it is expected to have the highest amount of diazinon in adjacent Abbandans. Kharajisha in Babolroud adjacency reveals more than others. When summer ends, diazinon application reduces gradually and its concentration falls in autumn similarly. Winter and spring have lower use in terms of crop pattern so its value is always lower in rivers and Abbandans as well.

Results show surface water concentration of diazinon is highest in summer months for rivers and abbandans. Diazinon is extremely low in abbandans from the initial sampling in April because its application had not yet started in the study area. After starting to use pesticide, the highest level of diazinon was detected in July compared to all other sampling times.

Conclusion

In most rice paddies in Mazandaran Province, diazinon is applied to control stem boring caterpillar of rice (*Chilo suppressalis*). It comprises about 23% of all used pesticides in Mazandaran and Golestan provinces (Abbasian et al., 2012). The existence of agricultural activities has a main role on surface and underground water resources pollution in Mazandaran province. The main problems are the high groundwater table in the region, and consumption of this water by local people as well as the anthropogenic side effects of water pollutants. Physical and chemical properties of the studied diazinon such as fumigation characteristic as well as the ecolo-gical conditions and soil type influence the reduction and eventual removal of the insecticides during the cultivation and harvest periods (Arjmandi et al., 2010). Retention time varies strongly in water, soil and biota. For example, a 2008 study by Ezemonye et al. showed decreasing order of occurrence of the diazinon as follows: fish > sediment > water. The fate of Diazinon in river depends on water outflow and degradation. In Tajan River, the si-mulated results showed Diazinon insecticide of about four months (Ahmadi, 2001). Two weeks after spraying, Diazinon reveals more than standard amount in common carp (*Cyprinus carpio*) and Chub (Leuciscus cephalus) in Qezel Ozan of Zanjan (Hamidi et al., 2012). Busher's study (2005) revealed that diazinon residue is more than admissible amount in rivers of Mond, Shahpour and Dalaky (Shayeghi et al., 2007).

Limited data indicated that yellowtail (*Seriola quinqueradiata*), a marine teleost, was 84X more sensitive to Diazinon than were 4 species of freshwater fishes, as judged by LC-50(48 h) values, and by its inability to biotransform Diazinon to nontoxic metabolites within one hour (Fujii and Asaka, 1982). Diazinon has not been detected in marine waters, but the potential exists for contamination of estuarine areas from agricultural and urban runoff (Eisler, 1986). Simulation of retention time of diazinon in river showed that highest concentration of diazinon affected fauna. The bioaccumulation of diazinon was studied in bluegill sunfish (*Lepomis macrochirus*) according to US EPA data requirements (Fackler, 1988). The steady state bioaccumulation factors were determined to be 470, 540 and 500X for the edible, non-edible and whole fish tissues respectively. Elimination of diazinon from these tissues was rapid, with half-life of between 1 and 3 days, indicative of rapid depuration.

It is very important to note that the rates of pesticide application in Mazandaran Province threaten both surface and underground ecosystems and this study can be a warning for potential decreases in ecosystem biodiversity. Water sampling from 10 shallow wells located in seven villages was carried out during summer and autumn of 2006. The results of the study showed the residue of Diazinon in groundwater of Mahmoud Abad area was 0.002 to 0.572 µg/l. In some samples the concen-tration of diazinon residues in water samples was higher than WHO maximum residue limits (0.µg/l) (Khazaei et al., 2010).

Obtained findings in this research reveal that farmers are applying huge amounts of pesticides and it shows the weakness of management and lack of an exact control and supervision on rate of pesticide consumption. Also it is impossible to forbid organ phosphorous pesticides usage in present condition, but this research emphasizes the importance of combinative (natural chemical) pest control to decrease the application of these kinds of pesticides. This controlling style can be a peaceful method between natural resources and agro ecosystems associated with sustainable development aspects.

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