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Investigation of the environmental distribution of endocrine disrupting materials in sewage sludge

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ABSTRACT

Today, as a result of the fast-growing industrialization, a high number of wastewater treatment plants, and the exceptional quantities of sewage sludge, disposal methods make environment even a more important topic. In Turkey, 600 million m^3 of wastewater in a year is treated by municipalities and 500,000 tonnes of dry matter originate from sludge. However, the methods used for the disposal of sewage sludge are usually land spreading and incineration. Although the methods of agricultural use of land and laying the sludge are limited by the European Union, there are empty quarries being used to fill the land. 17β-estradiol and estrone that are subject to the study are known as endocrine disrupters and being significant amounts at municipal wastewater treatment plant effluent and they remain in sewage sludge without any degradation. Hormones that are accumulated in the sludge, transfer into the soil with the rain and join the food chain. Experimental studies in the laboratory were carried out by simulating normal land conditions through advanced biological treatment plant sludge and with natural rainwater. In this study, three different rainfall amounts were selected for Istanbul province in April, May and October. The experimental set-up was dimensioned at the depth of 3 cm and 50 cm × 50 cm square filtrate rained by spray method. In addition, the amount of sludge being assigned to treatment laid hormones, endocrine disrupters have been identified in groundwater and soil mixing rates. This study was conducted to detect the amount of the hormone that can be mixed up with underground water, and it was detected that in the rains of April, May and October, a total of 69.02, 36.2, and 111.7 μg/m^2 for 17β-estradiol and 27.13, 10.1, and 45.8 μg/m^2 for estrone, respectively, dissolved from the sludge and transferred into underground water.

Keywords: Sewage sludge; Endocrine disrupting hormones; Rainfall; Land spreading

1. Introduction

In Turkey, as a result of the production increase, wastewater treatment facilities, andthe number and quality of treatment, plants continue to grow. As a result of this development, sewage sludge is a secondary pollution among the problems that need to be solved. 600 million m^3 of wastewater is treated by conventional treatment plants in a year [1]. According to

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the amounts of the treated wastewater, approximately 500,000 tonnes of sewage sludge per year results in secondary pollution. Although legal limitations stipulated by the European Union ban the use of sewage sludge agricultural or land spreading, charging empty mines is still preferred. In addition to spreading on land, agricultural use is also preferred for the disposal of sludge in Turkey. The amount of annual rainfall in Turkey should be taken into account and the amount of sludge contents, that can be transferred into the soil and underground water, should not be ignored. The subject of our study is identification the amount of endocrine disrupting hormones that can be soluble from soil and transferring groundwater via rainwater where the sewage sludge is being disposed as a method of spreading on land.

1.1. Definition of endocrine-disrupting substances

US Environmental Protection Agency (USEPA) defines endocrine disruptors (EDC) as follows: Organisms taken as external, natural or man-made, individual or population levels of chemicals which are reversible or irreversible effects. The European Union definition of an endocrine disrupter is; an exogenous substance that causes adverse health effects in an intact organism, or its progeny, secondary to changes in endocrine function [2].

1.2. Sources of endocrine-disrupting hormones

Many endocrine disrupters can accumulate in sediments due to the large and organic structures within the chain of biological life, after being discharged by wastewater. Furthermore, EDC can distribute around convection because of their stability and persistence. However, aquatic environment is affected by the endocrine-disrupters at most. EDC being accumulated in sediments adversely affect benthic organisms and fishes living in this environment [3,4]. Organisms who are nourished with high content of endocrine disruptive food are also adversely affected by these substances. In addition, endocrine disrupters may affect wildlife when they are present in the soil. It is reported that estrogenic hormones can be sourced to cattle grazing in the pasture, lasting storm water runoff, effluent water of fish farms, poultry house wastes, and chicken manures [5].

1.3. Side effects of endocrine disrupting hormones

Being exposed to endocrine disrupting hormones at a young age may cause unfavourable health effects. These effects may generally influence sexual development [6–8]. Researches show that endocrine disrupting hormones have adverse impacts on humans, as congenital malformations of reproductive organs lead to cancers and neurological disorders [9–13].

1.4. Recent studies

In a similar study, it is reported that 17α-estriol being one of the synthetic hormones is measured as 11 ng/g in treatment plant sewage sludge [14]. The present study identified estrogenic hormones in activated sludge and sewage sludge as estrone (E1), 17β-estradiol (E2) and 17α-estriol; in effluent water of plant there was no detection about these estrogenic hormones. As natural hormones, 17β-estradiol and estrone were adsorbed by sludge surface from water quickly in liquid phase [15]. In another study, 17β-estradiol concentrations and estrogenic activity in sludge dewatering processes were measured to be higher than the influent of treatment plant [16]. Endocrine-disrupting hormones were adsorbed to sewage sludge during treatment, and they were mostly detected in the filtrate phase, so they were re-pumped to the head of plant again and disposed with sewage sludge [17].

Researchers conducted a couple of studies and investigated the sludge applied on land. The first study was about a land near a channel fertilized with cow manure sludge applied for 3 months. The second study was conducted on the basin of rivers Rhine and Meuse, basins were left with the estimated amount of estrogen by animals living nearby [18]. In the first study, a total amount of one-year estrogenic hormones were fertilized for 3 months and measured to be as high as 150 ng/L. In the second study, Rhine and Meuse basins were fertilized with estrogenic hormones and measured as 75 and 140 ng/L, respectively, after 6 months from fertilization, 40 and 90 ng/L were measured, respectively. In another study, researchers fertilized the land with chicken manure and measured the initial concentrations of 17β-estradiol 20–2,530 ng/L, the highest level of leachate from the soil was observed as 675 ng/L [19]. In this study, leachate concentration of 17β-estradiol is proved depending on chicken manure application rate and frequency.

2. Materials and methods

2.1. Sample characterization

Atakoy Municipal Wastewater Treatment Plant sewage sludge which is located on the European coast of Istanbul province was selected for this study. Samples were collected in accordance with the sample conditions and they were characterized at Istanbul University Environmental Engineering Laboratory and TUBITAK Laboratory. Table 1 presents the parameters of sludge in the experiment setup.

2.2. Determination of moisture, dry matter, and ash

Volatile organic compounds were heated at 550–600˚C in an oven, percentage of moisture and dry matter content analysis were carried out at 1,05˚C in drying oven.

2.3. Estrogenic hormone analysis method in sludge

Samples were collected from Atakoy Advanced Biological Treatment Plant and kept in a freezer at −20˚C. Extraction method was applied to samples for measuring E1 and E2 quantities [20]. According to this method, samples were first freeze-dried and weighed 1 g, then treated with Na_2SO_4 and thoroughly put in a steel reactor. After adding 10 mL of acetone and 10 mL of methanol (1:1), samples were soaked for 5 min at 75˚C, under 103 bar pressure. Then they were evaporated until 1 mL sample remained, 1 mL sample was diluted to 100 mL with ultrapure distilled water. ELISA method was used to determine estrone and 17β-estradiol values.

2.4. Sewage sludge—rain water experiment setup

The study was conducted on the contents of the endocrine-disrupting hormones transferring from sewage sludge with rain water into the soil. After the stimulation of the transfer of rainfall into soil, 3 cm deep and 50 cm × 50 cm measured experimental setup was used (Fig. 1); sewage sludge was compressed using land applications. Experimental setup was filled with 33,000 g sewage sludge using land spreading applications in Istanbul and spray method raining was used over average total precipitations in April, May and October for Istanbul province with natural rain collected [21]. Rain didn't exceed the superficial rainwater stream, sludge provided all the rain filtered. Filtrate water was collected as samples and 17β-estradiol and estrone quantities were measured.

Table 1
Characteristics of sewage sludge

Humidity (%)	67.12
Ash (%)	62.03
C (%)	32.05
N (%)	3.87
H (%)	3.54
17β-estradiol (ng/g)	134
Estrone (ng/g)	59

3. Results and discussion

Characteristics of sewage sludge used in this study are presented in Table 1.

Natural rainwater was collected for simulating spray method originally. Rainfall data of Istanbul province was obtained from General Directorate of Meteorology (2014), average total rainfall data are presented in Table 2. According to classification of meteorological precipitation, rainfall is classified into six units (Table 3). By this classification, definite rainfall precipitations were sprayed through the experimental setup to explore the amount of rainfall which runoff and filtrate through the soil. To specify the precipitation class, strong, very strong, and heavy rainfall types were determined and thoroughly filtrated from experimental setup. For this reason, April, May and October rainfall statistics were selected for representative and similar values.

For April: very strong, 53.3 kg, for May: strong, 29.3 kg, for October: heavy, 67.9 kg rainfall precipitation were sprayed through the experimental setup. Figs. 2–4, show the 17β-estradiol concentrations of filtrate for April, May and October, whereas Figs. 5–7 show the estrone concentrations.

Total amount of 17β-estradiol concentration, dissolved from the sludge, decreased due to a change in precipitation of May. However, 17β-estradiol concentration increased with the time. Because of the high rainfall statistics of October, the dissolved 17β-estradiol concentration increased to higher levels.

Fig. 1. Land spreading simulation experimental setup.

Table 2
Istanbul province monthly average total rainfall (kg/m^2)

Month	Average rainfall (kg/m^2)
January	83.4
February	65.5
March	60.2
April	53.3
May	29.3
June	25.8
July	20.9
August	24.5
September	35.8
October	67.9
November	74.0
December	99.1

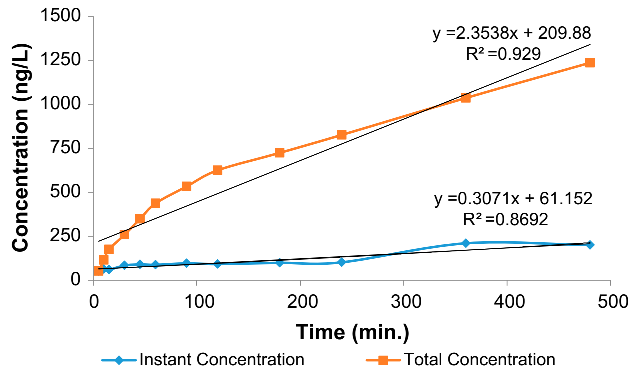

Fig. 3. Instant and total dissolved 17β-estradiol concentrations of filtrate of May.

Table 3
Classification of meteorological precipitation rainfall

Class	Rainfall intensity (mm)	Precipitation rate (mL/min)
Light precipitation	1–5	1.4–7
Medium precipitation	6–20	8–28
Strong rainfall	21–50	29–70
Very strong precipitation	51–75	71–104
Heavy rainfall	76–100	105–140
Excessive rainfall	>100	>140

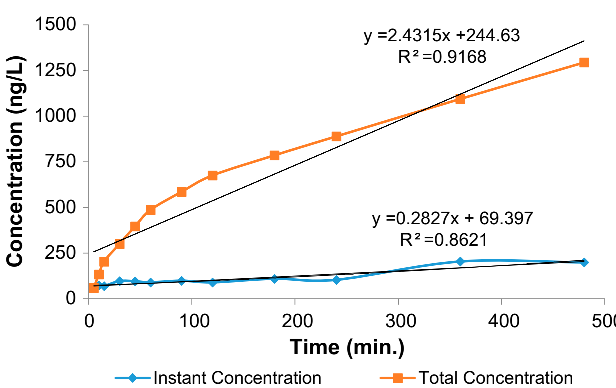

Fig. 2. Instant and total dissolved 17β-estradiol concentrations of filtrate of April.

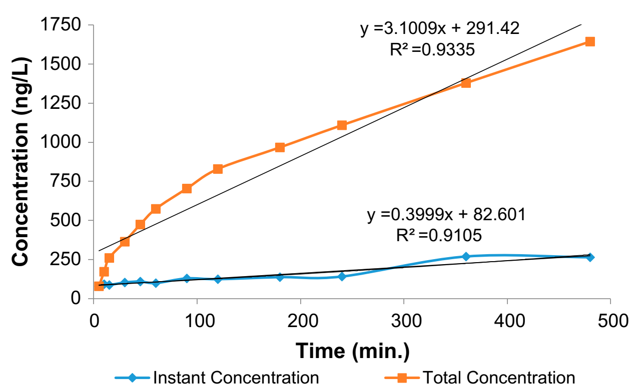

Fig. 4. Instant and total dissolved 17β-estradiol concentrations of filtrate of October.

During April, the dissolved estrone concentration increased disproportionately with time. During May, the dissolved estrone concentration was detected disproportionately, despite low precipitation statistics compared to April, the total concentration increased over time. The highest rainfall statistics for October was found to be the highest concentrations of estrone dissolved from sewage sludge.

In this study, negative environmental impacts of agricultural, recreational usage or storage removal methods of sewage sludge were simulated in laboratory. By spreading sewage sludge on land, runoff

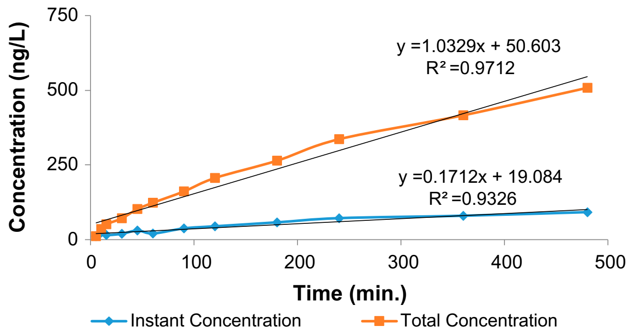

Fig. 5. Instant and total dissolved estrone concentrations of filtrate of April.

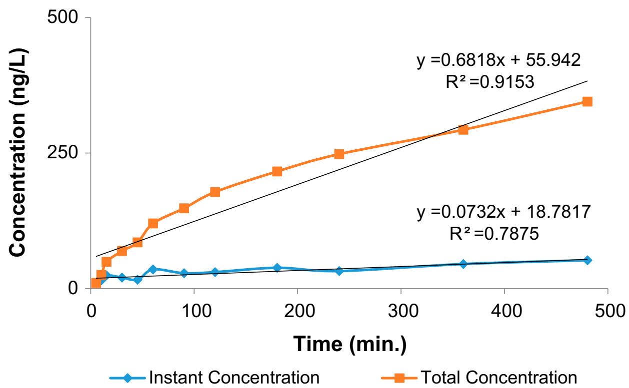

Fig. 6. Instant and total dissolved estrone concentrations of filtrate of May.

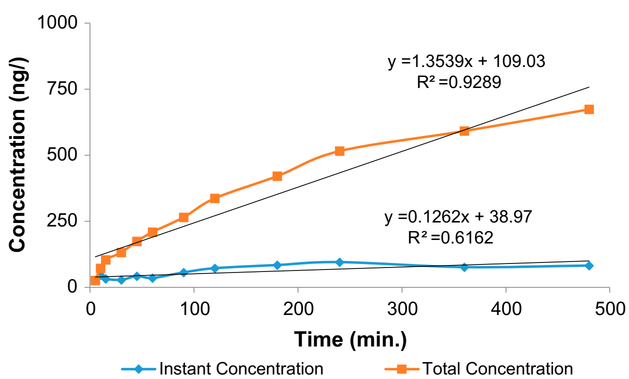

Fig. 7. Instant and total dissolved estrone concentrations of filtrate of October.

rainfall or rainfall filtrate through the soil was investigated and estrogenic concentrations were calculated. It is observed that, increasing amounts of precipitation shows higher dissolved estrogen concentrations. Block and Wösten [18] also reported in their study that estrogenic activity is not only filtrated through the soil but also reaches to surface water by runoff rainfall. The experimental setup was placed with a 33-kg sewage sludge according to land spreading conditions and 4,422 μg 17β-estradiol and 1947 μg estrone were calculated in sludge. At the end of the study, total dissolved 17β-estradiol concentrations were calculated for April as 69.02 $\mu g/m^2$, for May as 36.2 $\mu g/m^2$ and for October as 111.7 $\mu g/m^2$, whereas the total dissolved estrone concentrations were calculated for April as 27.13 $\mu g/m^2$; for May as 10.1 $\mu g/m^2$ and for October 45.8 $\mu g/m^2$. This study showed that estrogenic hormones in sewage sludge can be dissolved with rainfall through the groundwater. High estrogenic hormone concentrations in 1 m^2 sludge sample show that they can be dangerous for public health. Thus land spreading of sludge must be controlled and the European Union limitations should be applied.

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