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Industrial deinking paper sludge waste: toxicity risk and health effects assessment of heavy metals by USEtox model

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Abstract

Disposal of waste sludges produced in large amounts in the paper industry could generate significant environmental and health issues. One strategy to address them involves revalorization of deinking paper sludge (DPS) by reusing it as fertilizer. However, the possible human health risks associated with the use of DPS are still not well explored. The main objective of this report was to estimate DPS impacts on human toxicity. To achieve this goal, heavy metals analysis of the DPS waste (Cadmium; Cd, Copper; Cu; Molybdenum; Mo, Manganese; Mn; Lead; Pb; Cobalt; Co) was conducted. The assessment of human toxicity was performed by applying the UNEP/SETAC toxicity model *USEtox 2.0* to establish indicators that reflect the potential health damage of these chemicals when released into the environment.

Laboratory analysis, revealed a very low concentration of the DPS by the metallic contaminants (Cd, Cu, Mo, Mn, Pb, Co). According to the USEtox model results, these quantities will not lead to either carcinogenic or non-carcinogenic risks on human health even if there is a use of very high quantities of DPS. Indeed, the number of cases /t DPS emitted in agricultural soils didn't exceed 950.10^{-7} for the non-carcinogenic effect and $3.71.10^{-7}$ for the carcinogenic effect for Pb. For Mn and Co, we noticed no toxic effects (0 cases /t DPS emitted). Furthermore, we observed that Mo and Cu had very weak non-carcinogenic effects and led respectively to 445.10^{-7} and 56.10^{-7} cases /t DPS emitted. Regarding the effect of Cd toxicity, in order to have one case of this metal toxicity from DPS waste in our study, we had to use a very important quantity of DPS (≈ 2 821 680t). All these data emphasized on the absence of heath human toxicity risk after DPS waste industrial disposal, by ingestion or inhalation.

Keywords: Deinking paper sludge waste; Heavy metals; Human toxicity; Life Cycle Impact Assessment; USEtox model

1. Introduction

Identification and quantification of human health impacts associated to toxic substance utilization and emissions are thus critical for the development of sustainable technologies [1]. Industries have been identified as one of the most important sources of heavy metals in the environment as well as the first man-made environmental impacts [2]. Toxic

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(carcinogenic and no carcinogenic) contaminants (e.g, heavy metals) have become more prevalent, with the capacity to conduct to negative impacts and adverse effects on human health through the food chain [3- 4] **.**

Deinking Paper process have produced and continue to produce huge amounts of deinking sludge causing an environmental and economic impact on recycled paper mills [5].

Every year, 420 million tons of paper and cardboard are generated around the world. In fact, according to Bizjak *et al*[5], each year, 11 million tons of paper waste are disposed in european area of which, 70% appears from the generation of deinked recycled paper Moreover, around 70% of this waste appears from the generation of deinked recycled paper.

According to the Tunisian National Agency of Waste management TNAWM [6], waste production is increasing by 2.6 million tons per year, with paper and cardboard accounting for 8.6% of the total. According to the report National evaluation of indicators H2020/PAN Tunisia Kaabi *et al* [7], the creation of hazardous industrial waste by the industrial installation sector of paper, paperboard, and cardboard increased from 47 tons in 2002 to 91 tons in 2017. The majority of deinking paper sludge (DPS) produced across the world disposed in landfills, causing significant environmental damage and the loss of beneficial products present in the DPS [8]. Several studies conducted by Méndez et al [9]Marouani et al [10] and Vannucchi et al [8], have shown that DPS has biological and agronomic values and could contribute to the improving of the soil physical and chemical qualities.

Indeed, In order to generate recycled fiber from waste paper, ink, clay, coatings, and pollutants must be removed, resulting in large amounts of deinking paper sludge [11]. The paper industry waste is composed, in particular, of primary sludge rich in fibers from physical-chemical treatment, bark, ash, organic-rich secondary sludge from biological treatment and de-inking sludge from biological treatment and DPS from a deinking process[12]. DPS are high in cellulose fibers and carbonates, which can be pyrolyzed to develop adsorbents with high metal removal capacity [13,11]. According to Camberato *et al.*[14] and Warren [15]**,** the heavy metal content of deinking sludge is generally low. DPS may be more hazardous due to the high proportion of ink in the material. Today's printing inks include only these metals (Zn, Pb, Cu, Cr) due to advancements in ink technology [16]. Despite earlier inks that contained high levels of metals like Zn, Pb, Cu, Cr, Ni, Hg, Co, As, Se, and Sb.

As shown by Citeau [17], a soluble metal will pass into the water table or the plant, whereas in the case of insoluble ones it will remain in the soil. In fact, the solubility depends on several factors, in particular the soil acidity. Besides, the specific site factors like pedologicals and chemicals soils characteristics lead to bioavailability of these elements, thus the risk assessment [18].

Human exposure modelling is an important element that quantitatively links emissions to impacts. Several models have been used to report human toxicity indicators based on mechanistic methodologies accounting for fate, exposure and toxic effects providing cardinal impact measures [19, 20, 21]. In this context, different models were employed to calculate the effect of organic and inorganic chemical pollutants. In this investigation. In order to facilitate this study, USEtox model (version 2.0), which is available on the internet *(www.usetox.org)* have been considered. It determines both environmental impacts such as human toxicity and ecotoxicity, and was developed under the auspices of the United Nations Environmental Program (UNEP) and the American Society for Environmental Toxicology and Chemistry (SETAC) [22, 23, 24]. The compartments considered in this model are air, freshwater, sea, natural soil and agricultural soil [22].

USEtox™ is applicable in any comparative toxicity impact assessment (e.g. comparative risk/hazard assessment, ranking of chemicals according to their potential impact–comparative toxic benchmarking). Many researchers used the USEtox model in different fields as in freshwater ecotoxicity and human toxicity, on urban area, environmental aquatic ecosystems and on wastewater [24, 1, 23, 25, 26]. All these researches are based on mechanistic methodologies taking into account fate, exposure and toxic effects providing impact measures. As per to Rosenbaum *et al* [24], USEtox represents the best practice application as an interface between evolving science and a need for stability, parsimony, transparency and reliability.

According to author’s knowledge, there is no work to date on modeling the effect or the deinking paper sludge (DPS) assessment of on human toxicity using the USEtox model. Here, we addressed the following questions:

- Does DPS causes soil contamination?
- What are the effects of DPS on human toxicity using USEtox model.
- Is it reliable to use the USEtox model for the soil case?

2. Material and methods

2.1. DPS sampling

The current research was carried out on a Mediterranean Climate agriculture soil in Tunisia. Soil samples were taken at 2017 from the center (Enfidha) of Tunisia. The site is classified as BSh with 19.4°C and 320 millimeters of annual precipitation, according to the Koppen-Geiger climate classification.

In this research, the used DPS is an industrial deinking sludge, drained from wash water and discharged in the countryside (Figure1). The waste was acquired from the Tunisian paper industry (Tunisia Ouate) in March 2017. A random sample of the mixture of DPS waste was obtained from the point of discharge. The DPS samples were dried under vacuum in the open air, crushed, and sieved at 2 mm. For the metallic trace element (MTE) analyses, the sieved soil was maintained in boxes. Each sample was tested four times.

Figure 1 The deposed landfill

2.2. DPS heavy metal analysis

The analysis of heavy metals (Cd, Cu; Mo; Mn,; Pb and Co) in the biological amendment is carried out by a partial sludge digestion method [25]. As a first step, a hydrogen peroxide (H_2O_2) and nitric acid (HNO_3) digestion reagents are used with an Etnos One or Start Lab station control microwave heater with an internal temperature of 260-640°C and Easy control software installed, as well as an HPR1000/10s segmented high pressure rotor.. The heavy metals were determined using spectroscopic method by the ICP-OES apparatus.

2.3. Short description of the USEtox model

USEtox uses a matrix approach for multimedia modeling, which allows for the separation of fate, exposure, and ecotoxicity impacts when calculating the characterization factor (CF) (Figure2). The model incorporates current best practices, such as the inclusion of intermittent rain and Effect Factors (EF) based on substance toxicity across species. The USEtox database contains about 3.10^3organic compounds [23].

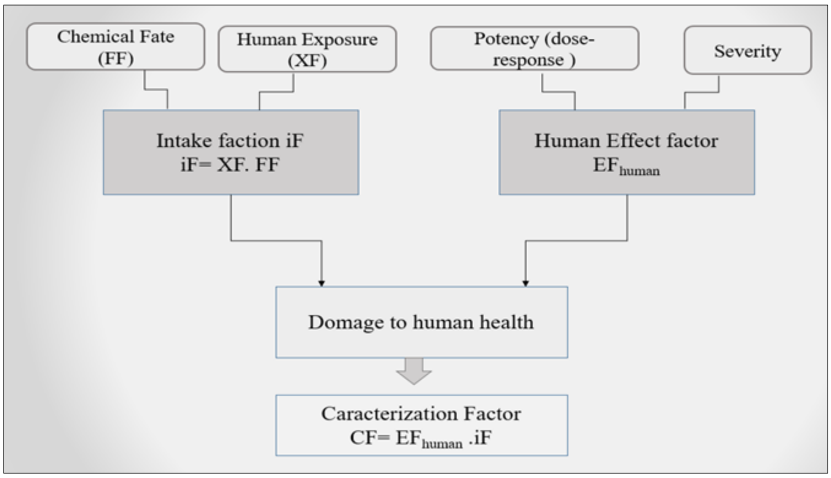

Figure 2 Short description of USEtox model Framework for human toxicity

To handle the USEtox model in practice, we should follow some critical steps. Firstly, we need to enrich its database by importing about 3500 organic and inorganic chemicals. After that, the substance-specific information for the chemical(s) under consideration has to be collected and stored in the "Substance Data" sheet [26]. Then, substances by name are selected in the initial window of the calculation setup wizard, when the first option "Run USEtox for 1-10 substances" is chosen [27]. Therefore, the choice of emission compartment and the environmental parameter is requested [28]. In the case of this study, the selected substances are Cd, Pb, Co, Mn, Mo and Cu emitted in agricultural land for the study area in North Africa. According to Westh *et al* [28] once the substances, emission compartments and parameters have been selected, the user is guided to the last window of the wizard: the result selection window (output). Finally, the "Run" sheet provides characterizations factors, fate factors, absorption fractions and effect factors of the chemical(s) under investigation [24].

In the Run worksheet, the main matrices with fate, exposure, intake fraction and effect factors are shown, followed by the characterization factor matrices [1] and [28]. The CF (the mid-point) for human toxicity is calculated by the continental and global characterization factors summation [22]. Indeed, the calculation is done through three steps:

- Fate in the environment where the distribution and degradation of each substance is modeled,
- Exposure, in which the human's exposure is modeled
- Effects, in which the inherent damage of the substance is investigated.

The cause-effect chain links are modeled using matrices populated with the corresponding factors for the successive steps of chemical fate factors (FF) in days, exposure factors (XF) in days^{-1} (human toxicity only) and effects factors (EF) in cases/kg ingestion human toxicity [24]. As a result, a set of characterization factors (CF) specific to the scale in cases / kg emitted is produced. In our study, the following formulas were used to calculate the adverse effects of the chemical substances on human health as well as on the ecosystem in relation to their concentration. For human toxicity the quantitative hazard representation is expressed by CF (cases / kg emitted):

$$CF = iF * EF \text{ ………………… } (1)$$

With (i) Effect Factor (EF_{human} (cases / kg intake)): the human health effects of the pollutant (EF expressed as carcinogenic or non-carcinogenic risk). (ii) iF (kg intake / kg emitted): (Intake Fraction); the fraction of the emitted mass that can affect the human population either by ingestion or inhalation.

The calculation of the midpoint results for each element was done according to the following equations and expressed in cases/kg emitted (1 person/kg emitted of TME):

$$\text{Midpoint}_{carc}(\text{TME}) = (EF_{carc} \times iF)_{Inhalation} + (EF_{carc} \times iF)_{Ingestion}$$

$$\text{Midpoint}_{non\ carc}(\text{TME}) = (EF_{non\ carc} \times iF)_{Inhalation} + (EF_{non\ carc} \times iF)_{Ingestion}$$

The midpoint results were obtained for each TME by the USEtox software in the general case and are presented in the table 2. This particular part of the risk analysis of the TME release

3. Results

3.1. TME content of deinking paper sludge (DPS)

The analysis of the MTE in the DPS revealed that values are below the thresholds established for the amendment*s* (Table 1).

However, it is crucial to assess if there are any human health risks linked to the release of DPS waste into the environment. The first-order impact (midpoint) of these MTEs (Cd, Pb, Mn, Co, Mo, Cu) on human health showed that the both carcinogenic and non-carcinogenic effects on humans (Table 2)

Table 1 TME content of dry matter in DPS and legal limits (mg.kg^{-1})

	Tunisian DPS (mg.kg^{-1})	*** US maximum allowable concentration in biosolids applied to soils**	****Residual sludge (Spanish legal limits)**
Cd	0.35 ±(0.036)	85	2500-4000
Mn	39.82 ±(0.42)		
Cu	35.16 ±(0.73)	4300	1000-1750
Mo	4.60 ±(0.38)	75	
Co	0.64 ±(0.019)		
Pb	2.24 ±(0.18)	840	750-1200
Ni		420	20-40
Zn		7500	1000-1500
Cr			300-400

* [17]** Legal limits established by the European Directive 86/278. The limits depend on the soil pH (minimum: pH < 7; maximum: pH > 7)[29]

Table 2 The results of the carcinogenic and non-carcinogenic effect factors (EF) (in cases/kg taken) and absorption faractions (iF) for TMEs under inhalation and ingestion obtained by USEtox

	Inhalation			**Ingestion**		
TME	**EF carcinogenic**	**EF non-carcinogenic**	**iF**	**EF carcinogenic**	**EF non-carcinogenic**	**iF TOTAL**
Cd	3.38	6.21	$2.21 .10^{-24}$	0.023	6.21	0.040
Pb	0.025	8.63	$2.28 .10^{-25}$	0.025	8.63	0.006
Mn	0	0	$1.38 .10^{-24}$	0	0	0.118
Co	0	0	$2.35 . 10^{-24}$	0	0	0.006
Mo	0	0.89	$4.98 .10^{-25}$	0	0.89	0.010
Cu	**0**	0.01	$2.80 . 10^{-25}$	0	0.01	0.017

TME: Total metallic elements, EF: effect factors, iF: intake factors, Cd: Cadmium, Cu: Copper, Mo: Molybdenum, Mo, Manganese: Mn, Pb: Lea, Co: Cobalt

3.2. Risks related to the release of DPS on human health

The risk associated with the DPS release into agricultural soils was calculated based on their MTE content (Table 2), even if they were found below the legal limitations established by the European Directive 86/278 [29].

As a result, we determined the DPS (midpoint) carcinogenic and non-carcinogenic effects per ton of de-inking sludge emitted. Furthermore, using the USEtox model's midpoint values (Figure3) and Excel (2007), we calculated the results in "cases / t DPS emitted." Table 3 summarizes the outcomes collected.

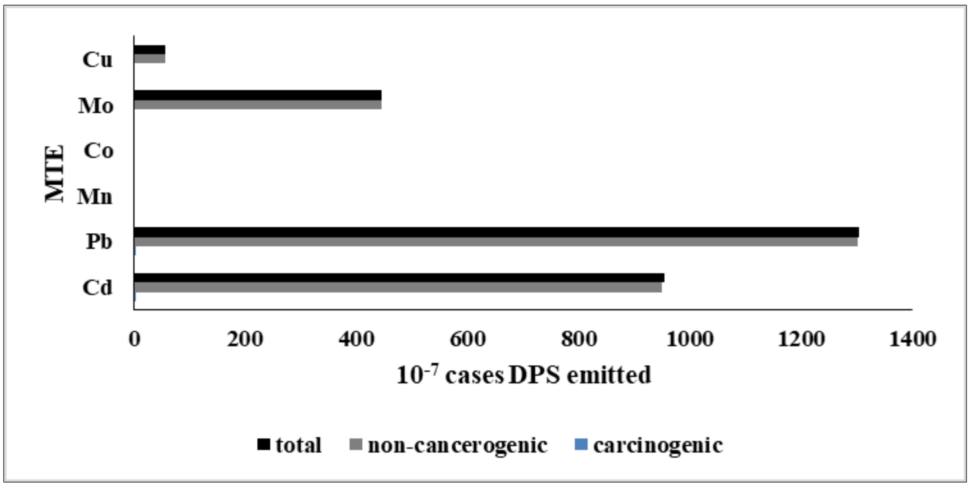

Figure 3 Midpoint results in cases /kg DPS emitted for carcinogenic and non-carcinogenic effects

We simulated the impacts of depositing one ton of DPS, and interestingly, no toxic effects on human health were identified. When emitted in agricultural soil, the number of cases/t of DPS didn t not exceed 950.10^{-7} and $3.71.10^{-7}$ for the non-carcinogenic and carcinogenic effects respectively and $3.71.10^{-7}$ for the carcinogenic effect in the case of copper. It should be emphasized that the trace metals Mn and Co have no harmful effects (0 instances / t of DPS released).

Table 3 The DPS release effects on human toxicity in agricultural soil

			Mettalics elements					
	Midpoint Units	**Toxicity**	**Cd**	**Pb**	**Mn**	**Co**	**Mo**	**Cu**
TME content	**mg.kg DPS**		0.35	2.24	39.82	0.64	4.6	35.16
USEtox	(case.kg issued TME)	Carc	0.001	16 .10^{-5}	0	0	0	0
		Non-carc	0.272	0.058	0	0	0.010	16 .10^{-5}
		Total	0.273	0.0583	0	0	0.010	16 .10^{-5}
Case.t DPS	10^{-7} case/t DPS issued	Carc	3.54	3.71	0	0	0	0
		Non-carc	950.82	130.87	0	0	445.06	56.2
		Total	954.37	1305.59	0	0	445.06	56.2
Quantity	t DPS per 1 case	Carc	2821681	2694553	0	0	0	0
		Non-carc	10518	7681	0	0	22469	177915
		Total	10478	7659	0	0	22469	177915

Carc : carcinogenic ; Non–carc : non-carcinogenic; t : ton, Cd: Cadmium, Cu: Copper, Mo: Molybdenum, Mo, Manganese: Mn, Pb: Lea, Co: Cobalt

According to the obtained data, the risks generated by the two elements Cd and Pb could occur into the agricultural soils only after emission about 2 821 680 t and 2 694 553 t, respectively. The current rate of production, this would take 966 years (8 t per day).

4. Discussion

Heavy metal pollution in agro ecosystems constitutes a serious environmental problem because of their toxicity, non-biodegradability, and particularly their high accumulation in soil and consequently in the food chain [4]. In recent decades, the used organic wastes in agriculture have gained increasing attention by researchers. Nonetheless, little investigations were explored of its impact on human health. The present investigation addressed the research question of how USEtox model could assess the risk of human toxicity.

Analysis of the TMEs in the DPS showed that the values of the different elements (e.g; Cu, Pb, Mn, Cd, Mo, and Co) are below the thresholds established for the amendments [29]. To confirm the impact of these elements on human risk, USEtox model was used as a suitable for the assessment of adverse consequences caused by the disposal of DPS waste either by ingestion and/or by inhalation in landfills. Rosenbaum *et al,* [26] explained that human exposure factors corresponding to the specific routes can be distinguished into direct inhalation or indirect ingestion exposure factors. In fact, the USEtox results confirm the studies of Flachier et al. [30], who showed that humans are exposed via inhalation due to exposure to soils contaminated with industrial waste.

The disposal site is situated outside of the city limits but near to an olive field. As a result, even if calcareous soil contributes to reducing the solubility and availability of some of the majority of metals, it is required to analyze the danger of contamination of food plants by various heavy metals [31]. Indeed, the daily food consumption (kg person. day-^{1}) of a Tunisian citizen for olive trees is 0.1 and 0.07 for adults and children, respectively. The risk of ingestion must be taken into account, especially for Cd and Pb [31]. The amount of metals absorbed by a human body (copper, zinc, lead, nickel and chromium) has a direct influence on its health. It can be acutely toxic as a result of cumulative exposure to a contaminated environment through their bioavailability in agricultural soils, or in the food chain [30]. Thus, there is a need for a certain health vigilance towards the presence of these elements in the soil [32].

In our investigation, DPS waste does not present any carcinogenic or non-carcinogenic risk to human health after inhalation or ingestion according to USEtox which confirms the results obtained by Marouani et al. [33]; Bailly et al.[34] and Camberato et al. [14], who argued on the fact that the content of TMEs in deinking sludge is generally low and has no risk of toxicity. According to Acero et al. [35], the amount of waste released impacts the calculation of human toxicity potential. However, according to Marouani *et al.* [33], the addition of DPS waste as an amendment, even at high doses (60t.ha^{-1}), did not result in a significant increase in soil MTE levels compared to the control, but improved some physical, biological, and chemical properties of the soil, resulting in better soil and environmental conservation. Adding to these results, beyond soil ingestion, the surrounding community may be exposed to health concerns from soil dust in their daily activities near the landfill [31].

The output of the midpoint model suggests that one case per kg of waste is emitted in nature. However, what is emitted is not only absorbed by humans, but its toxicity may have an impact on the food chain (e.g, plant, animal and man). Despite the results obtained in our data by the USEtox model, the latter remains having global averages [1]. Indeed, it takes into consideration as input only global and continental scale and general indoor environment setting (North, West, Est & Central Africa). As a result, allowed us to know and calculate the effects of some metals contained in the de-inking sludge on human toxicity (cases / kg emitted) and the general effects of these metals on emission to agricultural soil. Nevertheless, to assess better the effects of waste toxicity, several data on the study area and soil characteristics (pH, texture, moisture, plant, Temperature…)[4] must be considered as an input in the model account to properly quantify toxicity. In addition, Zang et al. [36], Wen-jia et al. [37] and Bejaoui et al.[31] showed that the atmospheric environment caused by the hazardous waste in the general industrial solid waste landfills should be also considered in the input of USEtox model.

Further researches are needed to establish the risks related to the direct human body contact with DPS waste during the handling in the analytical laboratory or in the field conditions.

5. Conclusion

Deinking paper sludge is an industrial product that poses an environmental risk. As a result, an assessment of its toxicity to human health is required. Here as a first step of the potential uses of USEtox model to assess the human risk of MTE released by DPS in the soil no toxic effects on human health were identified after application of one ton of DPS. More interestingly, based on the results given by the USEtox model, the MTE contents did not lead to either carcinogenic or non-carcinogenic risks on human health even at very high amount of DPS. Future research should also aim to identify the potential of other models to assess the human health risk after DPS application as well as in soil and plants.

Compliance with ethical standards

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Disclosure of conflict of interest

The authors declare no conflict of interest.

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