RESEARCH PAPER



The Impact of RDF Valorization on the Leachate Quality and on Emissions from Cement Kiln (Case Study of a Region in Morocco)

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ABSTRACT

Energy recovery is a sustainable method of municipal solid waste (MSW) management. The coincineration of refuse derived fuel (RDF) has shown several economic and environmental advantages. The objective of this research is to assess the impact of RDF recovery on leachate quality using leachate tests and calculation of greenhouse gases (GHG) reduction in the kilns of a cement plant. The qualitative results of the eluate show that there is an impact on leachate quality depending on the type of waste. The values of the chemical oxygen demand (COD), biological oxygen demand (BOD5), electrical conductivity and pH of the leachate from the raw waste after 120 hours of leaching are 29.33 gO_2/kg DM, 14.00 g O_2/kg DM, 4.27 ms/cm and 7.57. On the other hand, the values of the same quality parameters of the eluate generated by the waste without RDF are 19.33 g O_2/kg DM, 20.67 g O_2/kg DM, 2.77 ms/cm and 7.13; respectively. The calculation of GHG reduction shows that the substitution of 83,000 tonnes per year of petroleum coke by 15% of RDF (25,493 tonnes per year) can reduces 28,970 tCO₂ eq.

KEYWORD: Greenhouse gases; leaching test; Morocco; Municipal solid waste; Refuse-derived fuel.

INTRODUCTION

The quantities of household waste continue to increase due to rising population figures and improved living standards (Korai et al., 2017; Kassahun & Birara, 2018). The annual tonnage produced in urban areas was estimated at 1.3 million tons in 2012 which may double by the end of 2025 (Hoornweg and Bhada-Tata, 2012). In developing countries, landfilling remains the most common method of waste disposal. The main disadvantage of landfilling is that it generates large amounts of leachate, which is an effluent loaded with organic and mineral pollutants with high chemical oxygen demand (COD) and biological oxygen demand (BOD₅) values (Naveen and Malik, 2019; Han et al., 2016 & Shen et al., 2018). The characterization and degree of leachate pollution depend on the composition of the waste and the age of the

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landfill (Ziyang et al., 2009 & Bhalla et al., 2013). The majority of landfills have problems with leachate treatment due to their pollutant load (Tyrrel et al., 2002; Youcai et al., 2000 & Hussein et al., 2019). In order to assess the impacts of the waste, leachate tests are established. This technique was first used for hazardous waste and construction and demolition waste, (Kalbe et al., 2008 & Belevi and Baccini, 1989) and it was developed and used for household waste in order to get an idea about the soluble pollutants in the aqueous phase (Parodi et al., 2011).In order to minimize the impacts associated with landfilling, municipal solid waste (MSW) can be used as a source of thermal or electrical energy (Cucchiella et al., 2017). This mode shows several economic and environmental advantages (Scarlat et al., 2015 & Istrate et al., 2020). In particular, the refuse-derived fuel(RDF) process reduces the volume of waste, saves fossil fuels, incorporates residues into the composition of clinker and reduces CO₂ emissions from fossil fuels (El-Salmouny et al., 2020; Hemidat et al., 2019, Dondur et al., 2015; Scarlat, 2015; Ramachandra et al., 2018, Zhao et al., 2016, & Reza et al., 2013). The cement sector is a large consumer of fossil fuels (Schneider, 2015). Moreover, it is considered among the industries with the highest emissions of anthropogenic carbon dioxide (CO₂) at a share of 5-7% globally (Geng et al., 2019; Andres et al., 2012 & Kajaste and Hurme, 2016). The CO₂ generated by cement plants is due to the clinker manufacturing process and the incineration of fossil fuels (Geng et al., 2019). In order to achieve the 2030 sustainability goals, fossil fuels must be replaced by waste and other renewable energy sources (IEA, 2020). Studies have shown several economic and environmental benefits of co-incinerating alternative fuels at a 15% substitution rate in cement kilns (Kara, 2012; Çankaya et al., 2019 & Hemidat et al., 2019). In Morocco, the cement sector is undergoing an evolution in terms of cement production with twelve production plants that are distributed throughout the Kingdom with an annual production capacity of 21 million tons (APC, 2019). Hence, it is considered as the industrial sector with the highest energy consumption of about 30% of total energy produced (Fellaou and Bounahmidi, 2017). This study aims at assessing the environmental benefits of energy recovery from household waste in the Khenifra province of Morocco. First, a leachate test was conducted to evaluate the quality of the leachate generated by the landfilling of a raw waste and a waste without RDF. Then, calculations were made to evaluate the reduction of greenhouse gases (GHGs) emitted by the cement plant following the substitution of petroleum coke by 15% of RDF. This study was conducted in 2019 in the province of Khenifra in the Kingdom of Morocco.

MATERIALS AND METHODS

Waste management in the study area

The KhenifraProvince is located in the center of Morocco and is made up of 22 communities (Figure 1) with an estimated population of 315,552 inhabitants in 2020 (HCP, 2014). Since 2017, the large landfill site of Khenifra has been rehabilitated as a landfill and recovery center (Figure 1) to bury the waste of the 22 communities in a controlled manner. The estimated daily production of household waste in the province is 250 tons.

The scenario proposed in this study is the sorting of the combustible fractions to produce RDF for energy recovery in the kiln of the nearest cement plant in the province. The mass percentage of RDF is 28% (Ouigmane et al., 2017). The scenario is presented in Figure 2.



Figure 1. Geographic location of the study area and the cement plant of Khenifra Province in Morocco



Figure 2. The scenarios proposed for the present study

Leaching tests

In order to assess the impact of landfilling raw waste and waste without RDF on leachate quality, non-renewable leachate tests were carried out according to the AFNOR NF EN 12457-4 standard.

Samples preparation

The waste used in the test is taken from garbage bins in the city of Khenifra. Two types of samples were prepared; raw waste (RW) reconstituted according to the mass percentages of waste composition in Morocco (Ouigmane et al., 2018), and raw waste without RDF (RWWRDF) consisting of all other fractions except the dry ones. The samples were shredded into small fragments and mixed according to the standard AFNOR NF EN 12457-4. The mass composition of the two types of samples is shown in Table 1 (Ouigmane et al., 2017).

Fable 1.	Weight	percentage	of RW	and	RWWRDF
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Fraction (%)	RW	RWWRDF
Non combustible	72	100
Combustible (RDF)	28	0

Leaching parameters

The leaching tests were carried out with a liquid/solid (L/S) ratio of 10 at different durations without renewal of the liquid phase. Studies have shown that an L / S ratio of 10 should promote appropriate contact between the waste and the eluent (François et al., 2006 & Parodi et al., 2011). Distilled water was used as eluent (pH = 6.5; resistivity = 18.2 m Ω .cm). 100 g of the reconstituted and fragmented dry waste was mixed with 1 L of distilled water. The mixture was stirred on a stirring system. All experiments were performed at room temperature (20 °C) in glass vials. Leaching tests were performed in triplicate, with three vials removed after 120 hours every 24 hours (Figure 3).



Figure 3. Simplified schematic diagram of the experimental protocol of the leaching test without the renewable aqueous phase. *Analysis procedure*

Phase separation

The liquid was separated from the solid fraction by vacuum filtration (0.45 μ m). Then, all the solutions were stored in glass vials and kept at 4°C and analyses were performed rapidly

within 24 hours in triplicate. This step is critical since the COD can decrease by 9% after 15 days of storage (Parodiet al., 2011).

Analysis of the filtered liquid

After isolating the filtered leachate, several parameters including pH, electrical conductivity, COD and BOD5 were determined every 24 hours.

The pH of the leachate was used to identify the different degradation phases of the waste (Christensen et al., 1994). Indeed, this parameter governs the development or inhibition of certain degradation reactions taking place during the waste and leachate treatment process. It was determined using a pH meter according to the NF T90-008 standard. The electrical conductivity (EC) provides general information on the quantity of charged species present in the solid matrix. It is determined using a conductivity meter according to the NFT90-031 standard. The chemical oxygen demand (COD) gives a general idea of the oxidizable pollution in the leachate. It is a very useful parameter for the choice of the suitable type of treatment. It was determined according to NM 03.7.54 – 2013 standard. The biological oxygen demand in 5 days (BOD5) is used to study the biodegradable behavior of the generated leachate. Following the calculation of COD and BOD5, the COD/BOD5 ratio can be used as an indicator to help in the decision to choose a suitable treatment. BOD5 was determined according to the NM ISO 5815-2 (2012) standard.

Impact on carbon dioxide emissions

The aim of the second part of the research is the study of the influence of the substitution of petroleum coke by RDF in a cement kiln near the study area. The evaluation consists of calculating the reduction of gaseous emissions, including GHGs.

Calculations were made using the following formula:

$$E_{\text{reduction}} = E_{\text{no RDF}} - E_{\text{mix}}$$
(1)

Where: **E** _{reduction}: Reduction of GHG emissions in the cement plant (tCO₂eq/year); **E** _{no RDF}: GHG emission in the present cement plant if the pet coke (PC) is used at 100% (tCO₂eq/year) (Calculated according to the equation (2)); **E** _{mix}: GHG emissions in the cement plant if 15% of RDF is used as alternative fuel (tCO₂eq/year) (Calculated according to the equation (7)).

• Determination of E_{no RDF}

 $E_{no RDF}$ is calculated according to the following formula:

$$E_{\text{no RDF}} = E_{\text{PC, no RDF}} + E_{\text{landfill, no RDF}}$$
(2)

Where: **E** _{PC, no RDF}: GHGs emitted from PC if RDF is not used (tCO₂eq/year) determined according to the equation (3); **E** _{landfill, no RDF}: the methane emitted by the waste if the entire stream is buried in the landfill (tCO₂eq/year) (equation (5)).

$$E_{PC, no RDF} = m_{PC, no RDF} \times E_{t PC}$$
(3)

Where: $\mathbf{m}_{PC, no RDF}$: The amount of PC used in the cement plant without substitution by RDF (Tons/year), the amount of PC used each year is giving in table 2; $\mathbf{E}_{t PC}$ is the quantity of CO₂ emitted by the incineration of one tonne of pet coke (tCO₂eq/ t of PC) it is calculated according the equation (4)

 $E_{PC} = E_{PCMI} \times LCV_{PC}$

Where: $\mathbf{E}_{PC, MJ}$: CO₂ emitted from the incineration of PC to produce 1 MJ of heat (g CO₂/MJ) it's value is 98 g CO₂/MJ (EIA, 2019); **LCV**_{PC} : Lower calorific value of the PC used in the cement plant (MJ/kg) (30.4 MJ/kg).

The methane emitted by the waste if the entire stream is buried in the landfill is calculated according to the equation (5).

$$E_{\text{landfill, no RDF}} = m_{\text{MSW}} \times E_{\text{landfill, t MSW}}$$
(5)

Where: \mathbf{m}_{MSW} is the amount of municipal solid waste buried in landfill (Table 2); $\mathbf{E}_{landfill, t}$ MSW is the methane emitted by the waste if the entire stream is buried in the landfill (tCO₂eq/year) it is calculated according to the equation (6).

The methane generated by waste landfilling, it is calculated according to the Equation (6) (Kumar et al., 2004; Jensen and Pipatti, 2000; Komsilp et al., 2010 & Hosseini et al., 2018).

$$E_{\text{landfill, t MSW}} = (1t \times \text{MCF} \times \text{DOC} \times \text{DOC}_{\text{F}} \times \text{F} \times 16/12 - R_{\text{CH4}}) \times (1 - \text{OX})$$
(6)

Where;

MCF: Methane Correction Factor. According to the IPCC, the value used for landfills in developing countries is 0.4 (IPCC, 1995).

DOC: Biodegradable organic carbon. According to IPCC, the value taken is 0.19

DOC F: The fraction of biodegradable organic carbon converted into gas. According to IPCC, it is equal to 0.77.

F: Fraction of methane generated by the waste buried in the landfill, taken as 0.5 according to IPCC.

16/12: Conversion of C into CH₄

 \mathbf{R}_{CH4} : Recovered methane (Gg/year). Biogas recovery is not done in the study area so the value of this parameter is 0.

OX: Oxidation factor, the value taken by the IPCC is 0 (Pudasaini, 2014).

• Determination of E_{mix}

The value of E_{mix} is calculated according to the following equation:

$$E_{mix} = E_{PC, mix} + E_{RDF, mix} + E_{landfill, mix}$$
(7)

Where: **E** _{PC, mix} is the CO₂ emitted by the incineration of PC in the cement plant (tCO₂eq/ year) (calculated according to the equation (8)); **E** _{RDF,mix}: CO₂ emitted by the incineration of RDF in the cement plant (tCO₂eq/year) (calculated according to the equation (10)); **E** _{landfill}, mix: GHGs emitted from landfill if RDF is used as an alternative fuel (tCO₂eq/year) (calculated according to the equation (11)).

$$E_{PC, mix} = m_{PC, mix} \times E_{tPC}$$
(8)

Where: **m** _{PC, mix} is he quantity of PC used in the cement plant in the case of substitution by 15% of RDF (Ton/year) (Table 2); $\mathbf{E}_{t PC}$ is the mass of CO₂ emitted by the incineration of one tonne of pet coke (tCO₂eq/ t of PC) it is calculated according to the equation (9).

$$E_{tPC} = E_{PC, MJ} \times LCV_{PC}$$
(9)

(4)

Where: **E** $_{PC, MJ}$ is the mass of CO₂ emitted from the incineration of PCs to produce 1 MJ of heat (g CO₂/ MJ) it's value is 98 g CO₂/MJ (EIA, 2019); **LCV** $_{PC}$ is the lower calorific value of the PC used in the cement plant (30.4 MJ/kg)

$$E_{RDF, mix} = m_{RDF, mix} \times E_{t RDF}$$

Where: **m** _{RDF, mix} is the amount of RDF used in the cement plant (t/year) (Table 2); **E** _{t RDF} is the CO₂ emitted by the incineration of one tonne of RDF its value is 92 g CO₂ /MJ (Cong, 2017).

$$E_{\text{landfill, mix}} = m_{\text{MSW-RDF}} \times E_{\text{landfill, t MSW}}$$
(11)

Where: $\mathbf{m}_{MSW-RDF}$, is the amount of waste not recovered in RDF (t/year) (Table 2); $\mathbf{E}_{landfill, t}$ MSW is the methane emitted by one tonne of MSW if landfilled (tCO₂eq/ t of MSW) (calculated according to the following equation (6)).

Data used in the calculation

The lower calorific value (LCV) of petroleum coke used in the cement plant was 7500 Kcal/kg (30.4 MJ/kg). The LCV of RDF in the study area was 4453 Kcal/Kg (18.65 MJ/kg) as determined in the laboratory according to EN 15400 (2011). The mass of the RDF consumed in the cement plant and the estimated MSW used to produce RDF are presented in Table 2. The global warming potential of methane as a GHG is 25 times more than CO_2 (Brander et al., 2012).

Table 2. Various waste and PC data over 10 years in tonnes

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Parameter	2019	2020	2021	2022	2023	2024	2025	2026	2027	2028
m PC, no RDF	97000	97970	98940	99910	100880	101850	102820	103790	104760	105730
m _{MSW} ^b	191958	193890	195867	197889	199956	202069	204229	206436	208691	210994
m _{RDF}	25493	25765	26044	26329	26620	26917	27221	27531	27847	28169
m _{PC, mix}	83000	83830	84660	85490	86320	87150	87980	88810	89640	90470
$m_{MSW-RDF}$	166465	168125	169823	171560	173336	175152	177008	178905	180844	182825

^aCcalculated based on clinker production (1 million tonnes/year) in the first year and the energy required to produce one kilogram of clinker (3 MJ/Kg). (PCAFEAECE, 1975; Atmaca and Yumrutas., 2014)

^bEstimated by multiplying the population times the waste production ratio (0.78 kg/inhab/day in urban areas and 0.29 kg/inhab/day in rural areas) (Ouigmane et al., 2018).

RESULTS AND DISCUSSION

Leaching tests

pH and EC

The pH of the eluate generated by raw waste (RW) has increased from 6.67 after 24 hours to 7.57 after 120 hours (Figure 4). On the other hand, the pH of waste without RDF (RWWRDF), in the first hour of LT was relatively acidic (6.38) compared to the pH of the RW which evolved to 7.13 after 120 hours. The pH of the leachates found in various studies was reported to be neutral or alkaline (Yilmaz et al., 2010 & Magda and Gaber, 2014). The results of this study show that the pH is slightly alkaline which will adversely affect the solubility of heavy metals in the leachate (Costa et al., 2019). Thus, the pH is more acidic in the case of WRDF, which can promote chemical treatment of the leachate (Zhang et al., 2005 & Kurniawan et al., 2006). In the study by Sorlini et al. (2017), the authors found that residues from household waste

(10)

incineration have an acidic pH due to the presence of some elements such as aluminum. The advantage of RDF co-incineration is the incorporation of the ashes into the cement composition. The EC of the eluate generated by RW leaching was higher compared to the EC of the leachate generated by WRDF. After 24 hours of contact between the liquid phase and the waste, the EC was 3.5 ms/cm and 2.43 ms/cm for RW and RWWRDF; respectively (Figure5). After 120 hours, the EC value changed for both wastes, reaching 4.27 ms/cm for the RW and 2.77 ms/cm for the RWWRDF. In the study by Chantou (2012), the EC reported of LT of the raw waste was 3.8 ms/cm and 4.1 ms/cm after 24 and 120 hours; respectively. Since salinity is a source of mineral pollution and makes leachate treatment quite complicated (Magda and Gaber, 2014), the extraction of the combustible fractions of the waste can have a positive impact on the salinity level of the leachate generated.



Figure 4. pH monitoring results

Figure 5. EC monitoring results

COD and BOD5

The COD values were higher for RW with 19.33 g O_2 /kg DM after 24 hours of contact with the eluate. The COD of the leachate generated by the RWWRDF was 16.33 g O_2 /kg DM. The values of the two wastes changed every 24 hours (Figure 6). After 120 hours, analyses showed that the value reached 29.33 g O_2 /kg DM and 20.67 g O_2 /kg DM for the RW and RWWRDF; respectively. The COD found in the study of Parodi et al, (2011) after 24 hours of LT was 22 g O_2 /kg DM which was higher compared to the value found in the present study because of the composition of waste. The authors in the mentioned study used waste that was rich in combustible fractions (15.8% organic waste, 26.9% paper and cardboard, 12.7% paper and plastic, 4.9% textiles etc.). On the other hand, the BOD5 after 24 hours was 6.33 g O_2 /kg DM and 12 g O_2 /kg DM for RW and RWWRDF; respectively (Figure 7). After 120 hours, the BOD5 values evolved during the TL to reach 14 g O2/kg DM and19.33 g O2/kg DM for RW and RWWRDF; respectively.



Figure 6. COD monitoring results



The COD/BOD5 ratio plays an important role in the choice of effluent treatment. Widely used in wastewater treatment, this report evaluates the biodegradability of polluted water, i.e. the ability of organic matter to transform into mineral matter, admissible by the natural environment. For a predominantly domestic effluent, this ratio is generally between 2 and 3. For effluents from food processing industries, it is 1.5 to 2 order of magnitude less which reflects better biodegradability. Finally, when COD/BOD5 ratio is higher than 3, it reflects the contribution of an industrial effluent that is more or less difficult to biodegrade. As shown in Table 5, the COD/BOD ratio in the RW case was high compared to the WRDF. This indicates that the elimination of the RDF fractions made the effluent more biodegradable, which facilitated their treatment using a biological process that is less costly than combining biological and physico-chemical processes together (Abbas et al., 2009 & Costa et al., 2019). Thus, the COD/BOD5 ratio has decreased over time for both wastes. Indeed, studies have shown that the ratio of mature leachates is less than that of young leachates (Kjeldsen et al. 2002; Assou et al., 2016 & Renou et al., 2008).

Table 3. COD/BOD5 ratio in TL for both types of wastes										
Time		24 h	4 h 48 h			72h		96h	120h	
Type of Waste	RW	RWWRDF	RW	RWWRDF	RW	RWWRDF	RW	RWWRDF	RW	RWWRDF
COD/BOD5	3.05	1.36	2.95	1.26	2.85	1.35	2.47	1.24	2.10	1.07

CO₂reduction

The results of the calculation of the various equations were presented in the table in Appendix 1. From the analysis of the results (Figure 8), it can be seen that the value of the direct and indirect reduction of GHG emissions is significant. In the first year, the reduction is in the order of 28,970 tCO₂eq and it reaches 31,577 tCO₂eq in the 10thyear. Kara, (2012) found in his study that the use of RDF in a cement plant will directly reduce 5545 tCO₂eq with a substitution rate of 15% and a consumption of 18,921.6 tons/year of RDF. The study carried out by Hemidat et al, (2019) found that the substitution of petroleum coke by 15% RDF has directly reduced15, 890 tCO₂ eq. A study by Ecofys (2016) showed that the use of RDF as a pet coke substitute for clinker production in Greece reduced 168,000 tCO₂eqif the substitution rate was 7%. However, at 20% and 30% substitution rates, 480,000 tCO₂eq and 720,000

tCO2eq were reduced. In spite of the low emissions generated by Morocco, which does not exceed 0.2%, Morocco is committed towards all the international protocols which were declared recently in 2016 when it ratified the Paris agreement aiming at reducing GHG emissions. The country has decided to adopt a real roadmap for sustainable development which was documented under the National Sustainable Development Strategy (NSDS) (SNDD, 2017). The NSDS aims at achieving a vision of a green and inclusive economy in Morocco by 2030. The RDF development project intersects with the seven principles of the strategy and strongly meets the development objectives.



Figure 8. Evolution of CO₂ reduction over the years

CONCLUSION

The disposal of municipal solid waste is a sensitive step in the waste management process due to its adverse impact on the environment. This study was carried out in order to assess the environmental benefits related to the energy recovery of RDF produced from municipal solid waste in Morocco. The study showed that landfilling of waste without combustible fractions affects the quality of the leachate generated. The analysis of the pollution parameters in the effluents generated by both types of wastes showed a positive impact on the extraction of the dry fractions from the total waste stream. Thus, the COD/BOD5 ratio shows that the leachate from RWWRDF is biodegradable and can be treated by biological processes. A second advantage of RDF valorization lies in the direct and indirect minimization of greenhouse gas emissions. The case of the present study showed that the substitution of petroleum coke by 15% of RDF to produce one million tons of clinker can minimize 28970 tCO₂eq of emissions annually. The results of this study show that substituting fossil fuels with alternative fuels from household waste can minimize the pollutant load of leachate and reduce greenhouse gas emissions. Furthermore, this type of recovery increases the lifetime of landfills and reduces the volume of waste. Therefore, the next step in our work is to make a financial study on the use of this RDF as an alternative fuel in cement kiln in order to assess the sustainability of RDF valorization.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research

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Appendix :	The	results o	of the	calculation	of the	various	equations
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rependents. The results of the calculation of the various equations											
N° equation		2019	2020	2021	2022	2023	2024	2025	2026	2027	2028
1	E reduction	28970	29260	29549	29839	30129	30419	30708	30998	31288	31577
2	E no RDF	487643	492531	497464	502441	507461	512527	517640	522798	528003	533255
7	E _{mix}	458672	463271	467914	472601	477333	482109	486931	491800	496715	501678
3	E pc, no RDF	299524	302519	305514	308509	311505	314500	317495	320490	323486	326481
5	E landfill, no RDF	188119	190012	191950	193931	195957	198028	200144	202307	204517	206774
8	E PC, mix	247274	249746	252219	254692	257165	259637	262110	264583	267055	269528
9	E RDF, mix	23280	23513	23746	23978	24211	24444	24677	24910	25142	25375
10	E landfill, mix	163136	164763	166427	168129	169869	171649	173468	175327	177227	179169
4	E _{tPC}	2.98	2.98	2.98	2.98	2.98	2.98	2.98	2.98	2.98	2.98
6	E Landfill, t MSW	0.98	0.98	0.98	0.98	0.98	0.98	0.98	0.98	0.98	0.98



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