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## ASSESSMENT OF MERCURY EMISSIONS AND LOAD OF COAL AND PETROLEUM COKE BURNING AS ENERGY SOURCE IN CEMENT INDUSTRY

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### ABSTRACT

In Egypt, there is a power crisis. So search on varieties of power sources, especially in the highly consumed energy industries like cement is going in. Hence, Egypt began using coal as an energy source instead of Maseot and Natural Gas. However, coal combustion emits large amounts of air pollutants as mercury (Hg), sulfur content, carbon content and organic materials. The aim of the present study was to assess mercury emissions from coal burning cement industry at various cement companies that use different types of coal. This study was a descriptive cross-sectional survey that was conducted in Tourah, Helwan, and Arabian Cement Production settings that use coal as an energy source. Stack sampling of both vapor, and particulates' Hg was conducted according to the standard isokinetic method. Total stack Hg was then calculated. Helwan Cement plant (Coal) was of the highest Hg emissions [0.0008(0.011) mg/m<sup>3</sup>] followed by Tourah that uses petroleum coke [0.006(0.011) mg/m<sup>3</sup>] and Arabian (mixed coal) [0.003(0.006) mg/m<sup>3</sup>]. From the present study we concluded that Hg emissions of Helwan cement plant (petroleum coke) was higher than that of Tourah (Coal) and Arabian (mixed coal) ones. All plants were compliant with the Egyptian.

**Key words:** Cement industry; Coal; Energy Source, Mercury emission, Mineral coal, Petroleum coke.

## INTRODUCTION

Cement production consists of four main stages, including extraction and crushing of raw materials, pyroprocessing, as well as grinding and mixing (Gerbens-Leenes *et al.*, 2018). It is one of the most known energy-consuming industries that are not able to continue with the present situation of energy crises in Egypt (Abdulrahman and Huisingsh, 2018). So, the researchers in cement industry search for alternative non-traditional sources of power supply. Coal is one of the earth's energy sources, of which 50% of the carbonaceous materials are derived from ancient plants. The Indian and Chinese's cement industries use coal as a primary fuel (Verma and Kumar, 2017, Yan *et al.*, 2015).

Mercury is one of the heavy metals, which resulted as metal vapor or as metal compounds adsorbed on dust when coal is burned (Streets *et al.*, 2005, Contreras *et al.*, 2018). Minamata Convention October 2013 recommended reducing the mercury pollution in any ecological matrix (U.N., October 10-11, 2013). The Egyptian Environmental law number 9/2009 and its amended executive rule number 964/2015 set the emission limit of mercury produced from coal burning in cement industry as 0.05 mg/m<sup>3</sup> (Prime-Minister, 2015). German and Chinese's emission limits for both vapor and particulates Hg is 0.03 mg/m<sup>3</sup> (Sloss, 2012).

Health effects of Hg exposure vary according to its chemical form, whether it is elemental Hg (vapor), organic or inorganic Hg (particulates). Mercury vapor may lead to lymphocytic aneuploidy, neurological, reproductive, pulmonary, renal, muscular, and dermal symptoms

(Vahabzadeh and Balali-Mood, 2016). It can affect the foetus and young children's development (UN, 2013). Inorganic Hg may lead to kidney (Li *et al.*, 2015), and liver damage added to the neurobehavioral impacts (Yang *et al.*, 2016). Organic Hg is the most hazardous form, of which the main effect is neurological, in addition to the gastrointestinal, respiratory, kidney, liver, and dermal impacts (Risher *et al.*, 2002).

Global emission inventories 2010 indicated that cement production represents about 9% of the universal anthropogenic mercury emissions (UN, 2013). The mercury emission depends on the chemical composition of the burned coal, especially chlorine, bromine and other halogen contents (Streets *et al.*, 2005). US Environmental Protection Agency classified mercury compounds as hazardous air pollutants (HAPs) due to its toxicity, universal distribution of its sources, and long atmospheric residence time (Selin, 2009) (EPA, 2017). Worldwide studies commissioned by United Nations Environment Programme (UNEP) have confirmed the risks of global mercury emissions (UN, 2013).

In response to the Egyptian energy crisis, the Egyptian Government approved the use of coal fuel for both power generation and cement production, if certain conditions are met (Prime-Minister, 2015). Accordingly, it is necessary to evaluate the mercury emissions from coal-burning industries. So the aim of the present study was to assess mercury emissions and load from coal burning cement industry at various cement companies that use different types of coal.

## MATERIAL AND METHODS

This study was a descriptive cross-sectional survey that was conducted in Tourah, Helwan, and Arabian cement production settings that use coal as an energy source, and accepted to participate in the study. Mercury emissions (particulate or vapor) were sampled using Apex Iso-kinetic sampler – Model X5000 – Year 2012, according to the EPA standard methods. It was collected at a rate of 25 L/min for 120 minutes, the first 60 minutes for particulate sampling (six points from two ports) and second for mercury vapor at the same points and ports (US-EPA, 1996). The stack samples represented summer and winter seasons; samples was taken from two stacks built in two production line from each company with total 24 samples (12 samples for each season). Different companies (Tourah, Helwan, and Arabian cement) use different coal types, including petroleum coke, coal, and mixed coals (Hu and Cheng, 2016).

Mercury Samples were analysed using the standard method of ASTM D4185 and Dual amalgamation cold vapor atomic fluorescence spectrometric technique using CVAFS – Shimadzu Co. – 2001 – Japan according to the American Standard Technical Method (ASTM) D4185 (ASTM, 1996-2018). The volume of air sampled, the stack Hg concentration, and the Hg environmental load were calculated according to the Standards Method of EPA (EPA-OEE, 2007).

## RESULTS

In the present study, there were totally 36 samples of stack mercury vapor, 36 of stack mercury particulates. Stack mercury vapor, and particulates did not follow the normal distribution (Kolmogorov-Smirnov Test). Hence, the data were expressed as [median (interquartile range IQR)]. In the three companies, the concentrations of mercury emissions in its vapor form were lower than that in particulate ones.

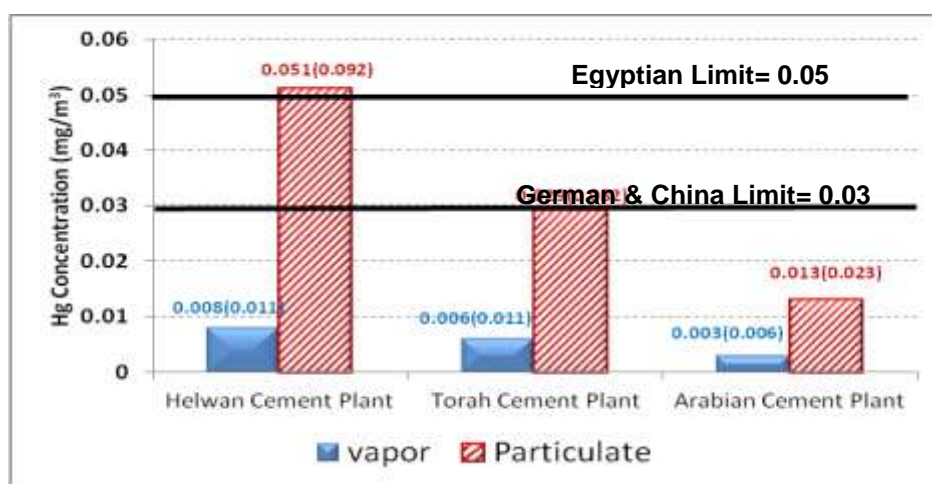
Hg vapor emissions in Helwan cement plant [0.008(0.011) mg/m<sup>3</sup>] was higher than that in Tourah [0.006(0.011) mg/m<sup>3</sup>] and Arabian [0.003(0.006) mg/m<sup>3</sup>] plants (Figure (1)). Kruskal-Wallis H Test disclosed its non-significant variation among the three plants. They were greatly lower than the Egyptian (0.05 mg/m<sup>3</sup>), German, and Chinese emission limits (0.03 mg/m<sup>3</sup>).

Next table shown the found analyses results in compare with the consumed burning coal and petroleum coke and amount of cement produced of each company.

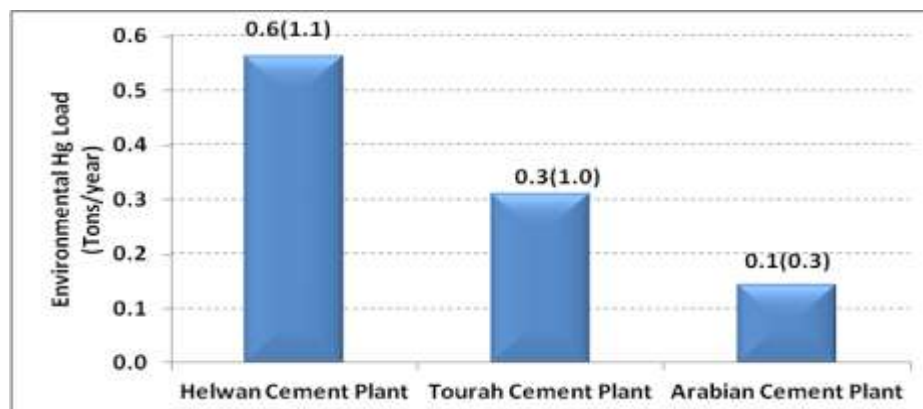
**Table (1):** Results of Mercury Load from coal and petroleum coke from three cement production plants

Company	Tourah	Helwan	Arabian
Average of Cement Production (Ton/Year)	30,000,000 (EEAA, 2015c)	5,395,753 (EEAA, 2015b)	4,650,000 (EEAA, 2015a)
Average of Fuel Consumed (Ton/Year)	177,343 (EEAA, 2015c)	144,895 (EEAA, 2015b)	100,129 (EEAA, 2015a)
Type of Fuel	Petroleum Coke	Coal	Mixed Coal
Mercury Content in the Fuel (mg/kg)	0.08 (Stoeppler, 2012)	0.16 (Stoeppler, 2012)	0.16 in 70% of fuel (as coal is 70% of the used fuel)
Hg Vapor Load Result (Ton/Year)	0.3	0.6	0.1
Hg Particulate Load Result (Ton/Year)	1	1.1	0.3
Total Hg Load (Ton/Year)	1.3	1.7	0.4

The environmental mercury load of Helwan Cement plant [0.6 (1.1) vapour (particulate)tons/year] was the highest followed by that of Tourah [0.3(1.0) vapour (particulate)tons/year] and Arabian [0.1(0.3)vapour (particulate)tons/year]. Kruskal-Wallis H Test revealed non-significant variation of mercury emission load among the three companies.



**Figure (1):** Levels of Mercury emissions (vapor and particulate) from Coal Stacks of different Cement Plants as compared with the Egypt, German, and China limits (2016-2017)



**Figure (2):** Environmental Load of Mercury Emissions from Coal Stacks of different Cement Plants (2016-2017)

## DISCUSSION

It is important to assess the mercury (Hg) emissions and the environmental Hg load from one of its main industrial sources in Egypt, which is cement productions, especially after allowing them to use coal as an energy source. Although Tourah Cement Plant had the highest cement production; Helwan Cement Plant had the greatest Hg emissions and annual Hg load followed by Tourah and Arabian. This may be attributed to the type of coal used in each company. Tourah uses petroleum coke, of which the Hg content (0.08 mg/kg coal) is half that of the mineral coal (0.16 mg/kg sample) that is used in Helwan Cement Plant (Stoeppler, 2012). The Arabian plant had the lowest Hg emissions and environmental load. This may be due to two main factors, including the advanced technology as well as the used kind of coal (mixed coal) (Osborne, 2013). Mixed coal is a mixture of mineral coal and organic wastes at a ratio 2:1. This ratio greatly reduces the Hg

contents of the mixed coal, and hence, mitigates the environmental Hg load.

Emissions of Hg vapor from the three companies were lower than the Egyptian limits of the Egyptian Prime Minister Decree no. 964-2015, annex no. 6, table no. 6 (Prime-Minister, 2015), as well as the German and Chinese limits (Sloss, 2012). Although US EPA considers Hg compounds of the hazardous air pollutants that may adversely affect public and environmental health (EPA, 2017), the Egyptian Environmental Law No 9-2009 and its executive regulation Number 964-2015 did not set standard emission limit for particulate Hg (Prime-Minister, 2015). In addition, China and Germany established the emission limits for both particulate and Hg vapor (Sloss, 2012).

In the present study, Hg emissions from the main stacks were higher than that from the by-pass ones. This can be interpreted based on the cement production line that has two stacks; the first is the main stack, which is the major line exhaust where all production and burning gases release from it. The other is the by-pass stack, which act as a secondary exhaust stack to avoid any damage in the production line, which may happen due to the huge amount of heat, excess CO<sub>2</sub>, release of particulates and the internal air pressure on the line's body. So, most emissions were logically from the main stacks' location (Osborne, 2013).



## **CONCLUSION**

From the present study we concluded that Hg emissions and environmental loads of Helwan cement plant was higher than that of Tourah and Arabian ones.

## **RECOMMENDATIONS**

The present study could not recommend using mixed instead of coal or petroleum; so the amount of mercury (Hg) emitted in air will be decrease as the result of the company emissions which use the mixed fuel is low.

## **RECOMMENDED FUTURE STUDIES**

It is suggested to conduct a study for evaluation of other emission products of mixed coal as compared to that of coal and petroleum coke to enable recommending the most suitable type. Moreover, it is recommended to study the annual mercury load from the three companies. In addition, it is proposed to apply a dispersion model to identify the ambient concentrations of Hg at different distances from the stacks.

## **ACKNOWLEDGMENT**

Great thanks are presented to the management of the three companies (Helwan, Tourha, and Arabian cement plants) for their permissions to conduct this study. The authors like to acknowledge the administrative support received from Institute of Environmental Studies and Research. They express great appreciation to the colleagues in the central laboratory, Egyptian Environmental Affairs Agency for their invaluable help to complete this work.

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## رصد تركيزات عنصر الزئبق في الهواء والمنبعه نتيجة حرق الفحم والفحم البترولي كمصدر للطاقة في صناعة الأسمنت

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### المستخلص

تبحث الدراسة تركيزات عنصر الزئبق المنبعث من حرق الفحم كمصدر للطاقة في صناعة الأسمنت بجمهورية مصر العربية.

الفحم هو واحد من مصادر الطاقة في كوكب الأرض، حيث يتميز بأنه وقود حفري قابل للاشتعال، حوالي ٥٠٪ من كتلة الفحم من مواد كربونية مشتقة من نباتات قديمة تعرضت للضغط وتعديل تركيب المواد العضوية الموجودة، حيث يختلف نوع الفحم حسب درجات تحول وتعديل المواد العضوية الموجودة في النباتات، بالإضافة إلى وجود مجموعة من الشوائب.

يستخدم الفحم في نطاق واسع اليوم في مصر كمصدر للطاقة بدلاً من البترول والغاز الطبيعية في فرع صناعة الاسمنت.

الزئبق هو واحد من المعادن الثقيلة التي تنتج كبخار أو مركب من مركبات الزئبق عندما يتم حرق الفحم، حيث تم وضع اتفاقية ميناماتا والتي وضعت خصيصاً للحد من التلوث بمركبات الزئبق أو ببخار الزئبق في أي منظومة بيئية وكذلك قانون البيئة رقم ٩٤/٤ ولائحته التنفيذية المعدلة بقرار رئيس مجلس الوزراء رقم ٢٠١٥/٩٦٤ والتي حددت الحد الأقصى لانبعاث أبخرة الزئبق التي تنتج من حرق الفحم في صناعة الأسمنت بنسبة ٠,٠٥ ملليجرام / م<sup>٣</sup>.

المواد والطرق المستخدمة

#### يمكن الحصول على الزئبق الذي تحمله جسيمات الأتربة من:

- المصادر الثابتة: وهي أكوام من عمل مصنع لإنتاج الأسمنت مع الفحم حرق كمصدر للطاقة.
- وحدة المصيدة الباردة (Cooled Trap).
- جهاز الامتصاص الذري - الجرافيتي.
- الطريقة القياسية المعتمدة من وكالة حماية البيئة الأمريكية والخاصة بسحب عينات الأتربة الكلية من المداخل من خلال جهاز مماثل لحركة الهواء داخل المدخنة ( EPA ISO-Kinetic ) (Methods no.5).
- الطريقة القياسية المعتمدة من وكالة حماية البيئة الأمريكية والخاصة بسحب عينات المعادن الثقيلة من المداخل من خلال جهاز مماثل لحركة الهواء داخل المدخنة ( EPA ISO-Kinetic ) (Methods no.29).
- الطريقة القياسية المعتمدة من وكالة حماية البيئة الأمريكية والخاصة بسحب عينات الأتربة الكلية من الهواء الخارجي لقياس تركيزات المعادن الثقيلة المدمصة على حبيبات الأتربة ( EPA Compendium Methods IO.5).
- تم أخذ عينات من الزئبق (الجسيمات أو البخار) باستخدام جهاز أبيكس إيزو-كينيتيك - موديل X5000 - عام ٢٠١٢، وفقاً للطرق القياسية ( EPA ISO-Kinetic Methods no.5 and Appendix I - no. 29 ) بمعدل ٢٥ لتر / دقيقة لمدة ١٢٠ دقيقة مقسمة إلى مرحلتين: أول ٦٠ دقيقة لأخذ عينات الجسيمات (٦ نقاط من منفذين في المدخنة - EPA ISO-Kinetic Methods no.5) و ٦٠ دقيقة ثانية لأخذ عينات بخار الزئبق (٦ نقاط من ٢ المنافذ في المدخنة - EPA ISO-Kinetic Methods no.29). تم أخذ العينات على مدار موسمي الصيف والشتاء. كما تم سحب العينات حسب خطوط الإنتاج داخل كل شركة حيث تم تحديد خطي إنتاج (الخط ١، والخط ٢) لكل شركة، وتم مراعاة أن الشركات الثلاث تستخدم ثلاثة أنواع من الفحم (فحم الكوك، والفحم البترولي، والمختلط) وذلك كله يأتي في إطار دراسة تأثير كل منها على انبعاثات الزئبق من المداخل.

تم هضم العينات التي تم جمعها من المداخل أو من الهواء الخارجي أولاً ثم حقنها في جهاز الامتصاص الذري، حيث تم استخدام جهاز من ماركة شيمادزو مشتملاً على وحدة المصيدة الباردة لتحليل تركيزات الزئبق الموجودة في محاليل العينات التي تم جمعها من المداخل سواء كانت عينات

فلاتر أترية أو محاليل مؤكسدة وكذلك العينات التي تم جمعها من الهواء الخارجي في المناطق المحيطة بمصانع الأسمنت الثلاثة طبقا لخطة العمل التي تم اعتمادها في خطة الدراسة.  
**النتائج ومناقشة النتائج:** بعد تحليل العينات واستقراء النتائج التي تم الحصول عليها وتحليلها طبقا للتحليل الإحصائي تبين الأتي:

- اتضح ان استخدام الفحم كمصدر للطاقة في صورة مختلطة مع مصدر طاقة آخر مثل النفايات البلدية يؤدي إلى تقليل كمية انبعاثات الزئبق التي تنتج من حرق الفحم الحجري النقي.
- سرعة الرياح واتجاهها تؤثر بشكل مباشر على تشتت الملوثات وتحديد تركيزات الزئبق المنبعثة من حرق الفحم في المصانع الثلاثة محل الدراسة.
- من خلال مقارنة تركيزات الزئبق المنبعث من حرق الفحم من مصانع الأسمنت تبين أن بعض النتائج قد تجاوزت الحدود القصوى المسموح بها في اللائحة التنفيذية الخاصة بقانون البيئة رقم ٤ لسنة ١٩٩٤ والتي صدر آخر تعديل لها بقرار من رئيس مجلس الوزراء رقم ٩٦٤ لسنة ٢٠١٥ حيث تجاوزت متوسطات تركيزات الزئبق المحمولة على الجسيمات الصلبة الكلية الناتجة من المداخن الرئيسية الحدود القصوى للقانون حيث أن الحدود القصوى للقانون هي ٠,٠٥ ملليجرام/م<sup>٣</sup> بينما كانت النتيجة التي ظهرت في متوسطات التحليل الإحصائي ٠,٠٨٧٥ ملليجرام/م<sup>٣</sup> بينما كانت أقصى تركيزات تم الحصول عليها في مصنع أسمنت حلوان حيث بلغت متوسطات تركيزات الزئبق في مداخن شركة أسمنت حلوان ٠,٠٥١٥ ملليجرام/م<sup>٣</sup>.