An Evaluation of the Scientific Evidence for an Alleged Threshold in the Relationship of Chlorine Input and Dioxin Output from Combustors

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By Pat Costner
Greenpeace International Science Unit
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Summary

Many scientific studies suggest that dioxin[a] formation by combustion systems can be abated by reducing or eliminating chlorine sources, such as polyvinyl chloride (PVC), in the materials to be combusted. Materials policies designed to reduce chlorine input and subsequent dioxin output from combustion systems have been implemented, acknowledged and/or proposed in various countries (e.g., Germany,[1] India,[2] the U.K.,[3] and the U.S.A.[4,5]), as well as in international fora such as the United Nations Environmental Programme[6] and the International Joint Commission on the Great Lakes.[7,8]

The broad utility of such materials policies was brought into question in a recent study by Wikstrom et al. (1996).[9] These scientists reported that decreasing the chlorine content of materials burned in their laboratory-scale combustor led to reduced dioxin emissions only when chlorine levels were 1 percent or greater. They went on to argue against materials policies as effective measures for dioxin abatement. Specifically, Wikstrom et al. (1996) contended that eliminating PVC from municipal waste streams would not achieve significant reductions in dioxin emissions from municipal waste combustors. They did not comment on the implications for total dioxin output.[b]

A reevaluation of the study by Wikstrom et al. (1996) suggests that their theory of a threshold in the chlorine/dioxin relationship at a chlorine concentration of 1 percent is not corroborated by their data. Given the variability in the low-level dioxin values obtained in this study, it seems probable that any apparent threshold is an analytical artifact. Moreover, whether the deviation observed is a threshold of detection or a threshold of formation, it does not occur at 1 percent chlorine. Further, it is not apparent that the seven studies cited by Wikstrom et al. (1996) as supporting their threshold theory can be readily interpreted to serve that purpose.

1.0 Introduction

[a] In this paper, the terms "dioxin" and "dioxins" are used to include all of the polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs).
[b] In this context, dioxin output is the quantity of dioxins released in all combustor residues, e.g., stack emissions, fly ash, bottom ash, scrubber water, etc.

Chlorine is the limiting element for dioxin formation in waste combustion systems. This circumstance suggests that dioxin output from such systems can be reduced and/or eliminated by controlling chlorine input through appropriately directed materials policies. This preventive strategy is supported by studies in which increased chlorine input has been accompanied by greater dioxin output. or specific fractions thereof (e.g., stack emissions and/or flyash). Such findings have been reported in studies with various combustion devices:

- Laboratory- and pilot-scale combustors: Gullett et al. (1997),[10] Pandompatam et al. (1997),[11] Kanters et al. (1996),[12] Halonen et al. (1995),[13] Burns (1993),[14] Halonen et al. (1993a),[15] Halonen et al. (1993b),[16] Wagner and Green (1993),[17] and Fangmark et al. (1993).[18]
- Full-scale combustors: Manninen et al. (1996),[19] Huotari and Vesterinen (1996),[20] Vesterinen and Flyktman (1996),[21] Moller et al. (1995),[22] Thomas and Spiro (1995),[23] Takeshita et al. (1992)[24], and Kopponen et al. (1992).[25]
- Home heating systems: Thuss et al. (1997).[26]

Materials policies can be especially important for dioxin abatement if wastes carry relatively high concentrations of one specific kind of chlorine donor or if one such chlorine donor contributes most of the chlorine in the wastes. For example, in Japan, the average PVC content of municipal solid waste has been reported as 12.2 percent, with a maximum of 25.3 percent.[27] In Denmark, municipal solid waste is estimated to contain approximately 0.6-0.7 percent PVC, which contributes about 67 percent of total chlorine input to incinerators in that country.[28]

Dioxin prevention is an important policy issue in all of the industrialized nations as well as many developing nations. Consequently, studies such as the study by Wikstrom et al. (1996) require detailed evaluation since they may have far-reaching influence on public health and economic policies.

2.0 A Review of the Study by Wikstrom et al. (1996)

The finding by Wikstrom et al. (1996) that dioxin emissions from their laboratory combustor increased with greater chlorine input is in agreement with many other studies, such as those cited above. However, these scientists also reported that the chlorine/dioxin relationship underwent a fundamental change when the chlorine content of the combusted material fell below a certain, critical level. In the abstract of their report, Wikstrom et al. (1996) identified this critical chlorine threshold as follows:[29]

> *"The results from this study indicate no correlation between the quantities of formed PCDDs/PCDFs ... in the combustion process and the level of chlorine in the fuel, when the chlorine level is below 1%. However, when the level of chlorine in the fuel exceeds 1%, an increased formation rate is noted."*

Based on these findings, Wikstrom et al. (1996) argued against chlorine materials policies aimed at reducing chlorine input to combustors by segregating PVC from the municipal waste stream:[30]

> *"Thus, the chlorine content in a normal MSW [municipal solid waste] is below the threshold value of 1% for an increase in PCDD/PCDF ... formation. ... The results of this study do not support the opinion that an elimination of only PVC will contribute to a considerable reduction of PCDD/PCDF emissions if the combustion process is well controlled, as in this study."*

It is important to note that Wikstrom et al. (1996) identified the critical threshold for chlorine content both as *"0.5 %"* as well as *"approximately 1 %"*:[31]

> *"...[N]o correlation is shown between PCDD/PCDF ... formation and chlorine content when the total chlorine amount in the fuel is 0.5% or lower. ... Figures 5 and 6 show that approximately 1% chlorine in these fuels is a threshold value for the formation of excess PCDD/PCDF ... during combustion."*

The average dioxin concentrations measured at each chlorine level, as shown in Tables 1 and 4 of the paper by Wikstrom et al., were subjected to minor corrections in addition, as indicated below in Table 1. When these data are presented as a scatter plot, Figure 1, it is evident that dioxin emissions continued to decline as the chlorine content fell to 0.56 percent.

Table 1: I-TEQ (in ng/Nm3) in Flue Gas for All Samples from Each Fuel (Taken from Table 4 of Wikstrom et al. (1996))

%Cl	Experiment 1		Experiment 2		Experiment 3		Total, by Wikstrom	Total, Corrected
	I	II	I	II	I	II		
0.12	0.15		0.30	0.22	0.08	0.05	0.16	na
0.20	0.05	0.08	0.18	0.15	0.10	0.10	0.11	na
0.46	0.21	0.20	0.21	0.29	0.24	0.23	0.23	na
0.63	0.17	0.16	0.17	0.14	0.28	0.15	0.15	0.18
0.56	0.08	0.07	0.12	0.26	0.24	0.20	0.15	0.16
0.84	0.21	0.30	0.64	0.67			0.46	na
1.72	1.23	1.55	1.28	1.32	1.47	0.72	1.26	na

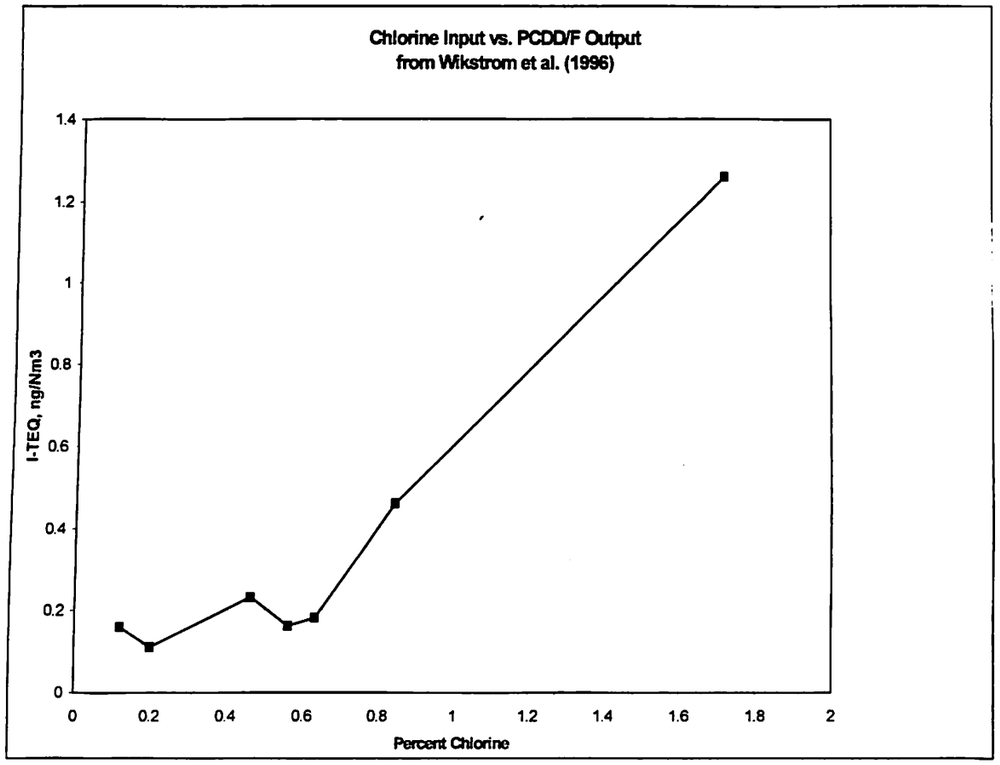

Figure 1: Experimental Data (Corrected) from Wikstrom et al. (1996)

Several inferences can be drawn from the data obtained by Wikstrom et al. (1996). For example, since the uncertainties of dioxin values increase at very low dioxin concentrations, the apparent discontinuities in the correlation of chlorine input and dioxin output at chlorine levels below 0.56 percent may represent a threshold of detection rather than a threshold of formation. Also, the data may be indicative of a greater than linear, or exponential, relationship between chlorine input and dioxin emissions, similar to that reported earlier by De Fre and Rymen (1989).[32] In any case, the data presented by Wikstrom et al. (1996) do not appear to support the conclusion that the positive correlation between chlorine input and dioxin emissions ceases to exist when the chlorine content falls below 1 percent..

3.0 Other Studies Cited as Supporting the Threshold Theory

In their paper, Wikstrom et al. (1966) cited seven other studies in support of their hypothesis that there is a threshold below which the positive relationship between chlorine input and dioxin emissions no longer exists:[33]

> *"Seven of these studies found an increase in the PCDDs/PCDFs formation when the chlorine content in the fuel exceeds a threshold value at 1-2% ..."*

The seven studies, in order of their appearance in Wikstrom et al. (1996), are as follows:

Halonen et al. (1993);[34]
Mattila et al. (1992);[35]
Takeshita and Akimoto (1989);[36]
Wagner and Green (1993);[37]
Lenoir et al. (1991) (cited as *"Leonir et al. (1991)"*;
Ruuskanen et al. (1994);[38] and
Frankenhaeuser et al. (1993).[39]

As discussed below, the data and conclusions presented by the authors of these studies do not corroborate the above statement by Wikstrom et al. (1996). I.e., there is little if any indication that any of these studies found a threshold at 1-2 percent chlorine. It is important to note, however, that most can be said to lend support to materials policies as effective dioxin abatement measures.

3.1 Halonen, I., Tarhanen, J., Kopsa, T., Palonen, J., Vilokki, and H. Ruuskanen, J. 1993. Formation of polychlorinated dioxins and dibenzofurans in incineration of refuse derived fuel and biosludge. Chemosphere 26 (10): 1869-1880.

Wikstrom et al. (1996) included this study by Halonen et al. (1993) among those cited as supporting their hypothesis of a threshold of 1 percent chlorine, as follows:[40]

> *"The results from our study are in good agreement with the data presented by Halonen et al. (18), Mattila et al. (21), and Leonir et al. (26) (sic). All these studies indicate that approximately 1% chlorine in the fuel represents a threshold value for increased PCDDs/PCDFs formation."*

The Swedish scientists pointed out that *"Halonen et al. (18) combusted three fuels with different chlorine levels (0.2, 0.35, and 2.1%)."* However, Halonen et al. (1993) seem to have given two differing values for the highest chlorine concentration. In a graph, the highest chlorine content -- that of refuse derived fuel (RDF) -- is given as 2.1 percent, while, in the text, it is given as 1.0 percent, as shown in the following excerpt:[41]

> *"The total chlorine content of RDF fuel was 1.0 wt-%. ... The chlorine content of the biosludge/wood chips mixture was 0.2 wt-% ... The corresponding values of the biosludge/bark mixture were 0.35 wt-% ..."*

Fortunately, Halonen et al. (1993) also characterized the chlorine input rates for the three fuels as follows: biosludge/chips, 0.16 mg/s; biosludge/bark, 0.21 mg/s; and RDF, 0.58 mg/s. Correlation of these values with those for percent

chlorine suggests strongly that the chlorine content of the RDF was 1.0 percent, as reported in the text, rather than 2.1 percent, as given on the graph and relied upon by Wikstrom et al. (1996).

Wikstrom et al. (1996) also described the findings by Halonen et al. (1993) as follows:

> *"No differences in the emission levels of PCDDs/PCDFs from the fuels with 0.2 and 0.35% chlorine were observed. About three times higher emission of PCDDs/PCDFs was observed from the fuel with the highest chlorine level."*

Somewhat more exactly, Figure 2 in the paper by Halonen et al. (1993) shows considerable variation among the samples that were taken at three locations. As shown by the data in that figure, which are excerpted and re-graphed in Figure 2 below, the dioxin concentrations in samples taken after the hot cyclone (AHC) exhibit a linear relationship with chlorine input ranging from 0.58 mg/s to 0.21 mg/s. If the data taken at the other two sampling locations -- before the baghouse (BBH) and after the baghouse (ABH) -- can be said to exhibit a threshold, such threshold occurs at some undetermined point between 0.58 mg/s and 0.21 mg/s, or, respectively, 1 percent and 0.35 percent chlorine.

It is apparent that the results of this study do not entirely agree with the characterization offered by Wikstrom et al. Rather, Halonen et al. concluded as follows:[42]

> *"PCDD/PCDF formation at 850 C was observed to depend on chlorine content in the fuel. ... The results of the three fuel mixture incineration tests (fig. 2) show that increased chlorine content in the fuel produced higher PCDD/PCDF concentrations formed at 850 C (AHC), but similar correlation at 200 C (ABH) was not seen in combustion tests of biosludge. .. It is obvious that variables other than the chlorine and PCDD/PCDF input contents in the fuel have an influence on formed PCDD/PCDF levels. ... The chlorine content of fuel materials had the most effect on PCDD/PCDFs formation at the high temperature region of the flue gas. When the flue gas temperature decreased the correlation of the chlorine content in fuel with the formation of PCDD/PCDF compounds disappeared."*

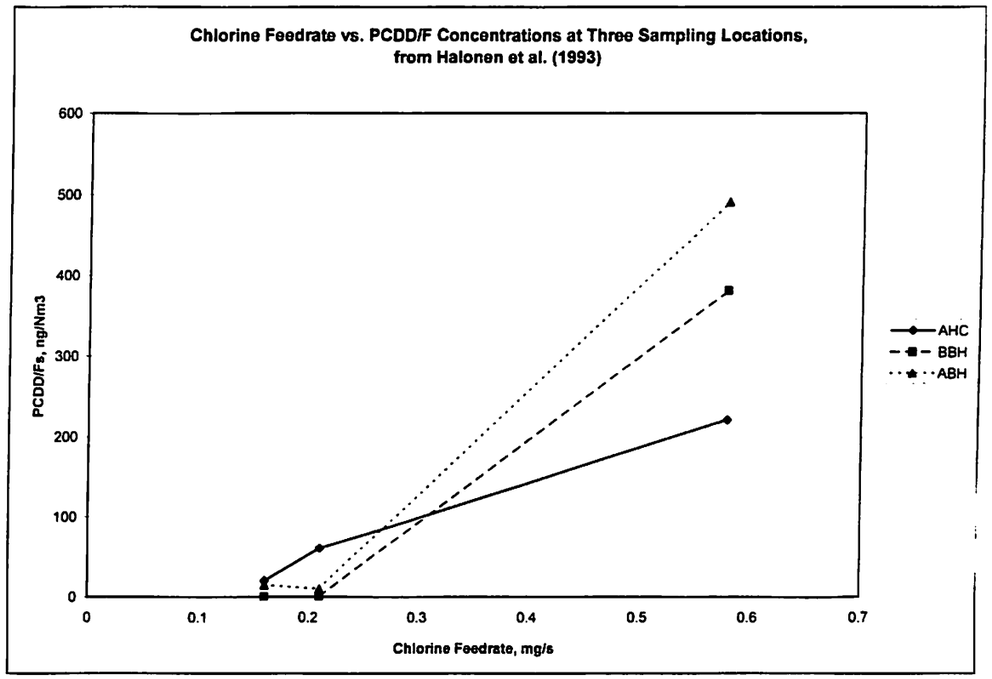

Figure 2: Experimental Data from Halonen et al. (1993).

In summary, the study by Halonen et al. (1993) does not support the conclusion by Wikstrom et al. (1996) that chlorine input influences dioxin output only when chlorine content exceeds 1 percent. It does, however, lend support to the usefulness of a chlorine materials policy as an effective dioxin abatement strategy.

3.2 Mattila, H., Virtanen, T., Vartiainen, T., Ruuskanen, J. 1992. Emissions of polychlorinated dibenzo-p-dioxins and dibenzofurans in flue gas from co-combustion of mixed plastic with coal and bark. Chemosphere 25 (11):1599-1609.

Wikstrom et al. included this study by Mattila et al. (1992) among those said to support the existence of a 1 percent threshold for a positive relationship between chlorine input. The Swedish scientists further described the outcome of the study by Mattila et al. (1992) as follows: [43]

> *"Mattila et al. (21) ... A considerable increase in the formation of PCDDs/PCDFs was noticed between the experiment with low PVC loaded (0.4% Cl and high (1.2% Cl.)"*

As shown below in Figure 3, the data presented by Mattila et al. (1992) support the above statement. However, these data do not support the existence of a threshold at 1-2% chlorine. Rather, they show a rapid decline in dioxin

emissions as the chlorine content drops from 1.2 to 0.4 percent, followed by a slower decline as chlorine content decreases from 0.4 to 0.04 percent.[44]

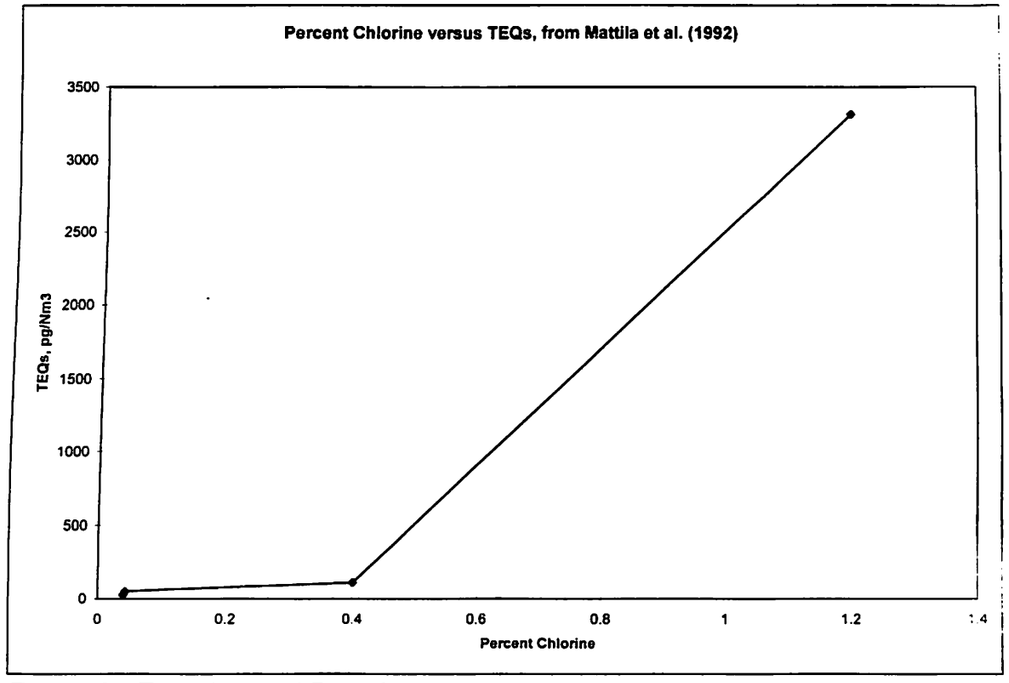

Figure 3: Experimental Data from Mattila et al. (1992).

It is evident that these data do not support the hypothesis that there is a threshold below which chlorine input and dioxin output no longer correlate. Rather, the results obtained by Mattila et al. (1992) suggest the possibility of an exponential relationship between chlorine input and dioxin output.

Mattila et al. (1992) summarized the results of their study, which was carried out with a full-scale combustor, as follows:[45]

> *"Our results prove the evidence for conversion of chlorinated plastic material to polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). ... the total amount of PCDD/Fs correlate closely with the formed HCl concentration in tests 1 to 3, but in test 4 the yield of PCDD/Fs was ten times higher than expected. ... This full scale study showed that the formation of PCDDs and PCDFs is connected with the total chlorine content in the fuel mixtures. The results give a strong support to the PCDD/F formation mechanism involving chlorinated plastics as the identified chlorine source. The second major implication of the PCDD/F formation is its relationship to the concentration of HCl formed during combustion."*

3.3 Takeshita, R., and Akimoto, Y. 1989. Control of PCDD and PCDF formation in fluidized bed incinerators. Chemosphere 19 (1-6): 345-352.

This study was among the seven studies described by Wikstrom et al. (1996) as supporting their hypothesis that there is a threshold at 1 percent chlorine. Takeshita and Akimoto (1989) described their study as follows:

> *"Therefore, the object of this investigation was to establish how control of hydrogen chloride in flue gas might decrease the formation of PCDDs and PCDFs in the incineration. ... the relationship between levels of formed PCDDs and PCDFs and levels of hydrogen chloride formed under combustion was investigated. ...levels of PCDDs in the EP outlet gases tended to decrease in proportion to the decrease in levels of hydrogen chloride in the flue gases. ... At higher levels of hydrogen chloride in the gases, PCDFs were increasingly formed."*

In this paper, the researchers present no data describing the chlorine content of the municipal solid waste that was burned in a full-scale, fluidized bed combustor during the addition of incremental quantities of dolomite. Values for HCl concentrations and dioxin output (in gases, particulates and ash from the electrostatic precipitator as well as boiler ash) were estimated from graphs presented in this paper and presented below in Figure 4. As shown, these data exhibit a linear relationship between HCl and dioxins.

In summary, neither the type of data collected in this study nor the trend shown by these data support the hypothesis of a threshold at 1 percent chlorine. They do suggest, however, that a chlorine materials policy can be an effective measure for dioxin abatement.

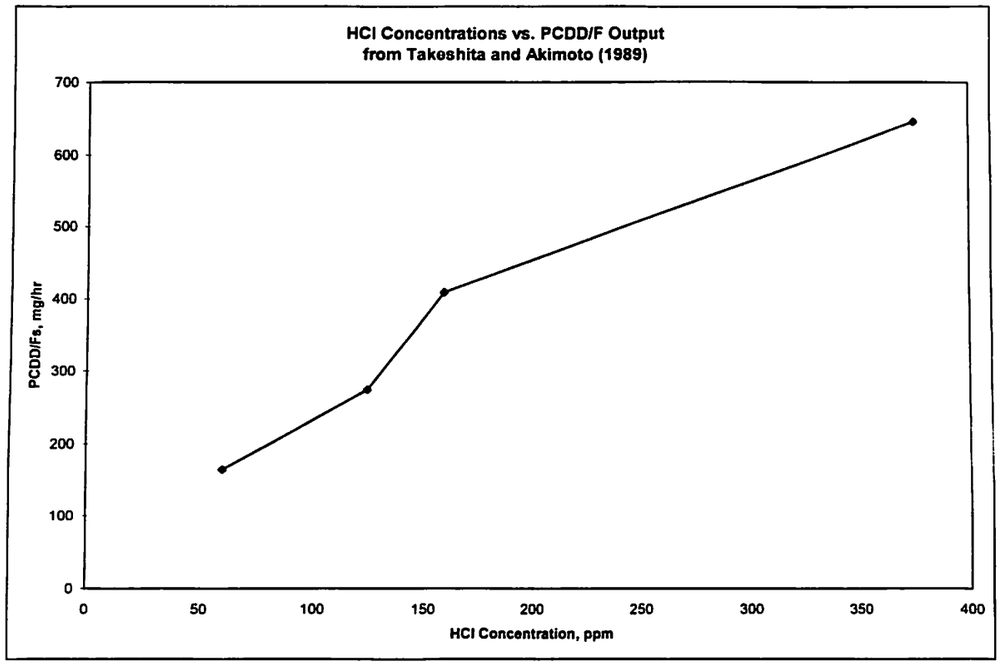

Figure 4: Experimental Data from Takeshita and Akimoto (1989)

3.4 Wagner, J., and Green, A. 1993. Correlation of chlorinated organic compound emissions from incineration with chlorinated organic input. Chemosphere 26 (11): 2039-2054.

This study was also cited by Wikstrom et al. (1996) as supporting their hypothesis of a threshold for the chlorine/dioxin relationship. Wagner and Green (1993) examined the relationship of chlorine input and volatile organic compounds including chlorobenzene and dichlorobenzene, which are among the chemicals that are regarded as indicators for dioxin. They combusted non-hazardous waste to which varying quantities of PVC were added to achieve chlorine contents up to approximately 2.75 percent. These U.S.-based scientists concluded as follows:

"We find several statistically significant relationships between HCl emissions (a surrogate for PVC in the waste) and the emissions of a number of chlorinated organic compounds. ...These results, contrary to the prevailing opinion [1], lead to the physically reasonable conclusion that decreases in the levels of organically bound chlorine in the input leads to decreases in chlorinated organic emissions. ... In final summary the CCTL's experimental, phenomenological, and theoretical studies of toxic emissions from incineration all support the physically intuitive hypothesis that reduction of chlorinated plastics in the input waste stream results in reduction of aromatic chlorinated organic emissions

[21]. ... While the CCTL's measurements have been limited to volatiles and light semi-volatiles these results are expected to apply to other chlorinated aromatic hydrocarbons emissions such as phenols, dioxins, and furans, which we have not measured. A number of other publications support a PVC-PCDD association [22-26]. Bulley has also found experimentally that reduction of PVC input leads to reduced chlorinated dioxins and furans [27]. Thus we are convinced that, when all other factors are held constant, there is a direct correlation between input PVC and output PCDD/PCDF and that it is purposeful to reduce chlorinated plastics inputs to incinerators"

Data describing HCl and dichlorobenzene were excerpted from this report and presented below in Figure 5. It is not readily evident that these data from Wagner and Green (1993) support the threshold hypothesis either by the type of data collected or their trends.

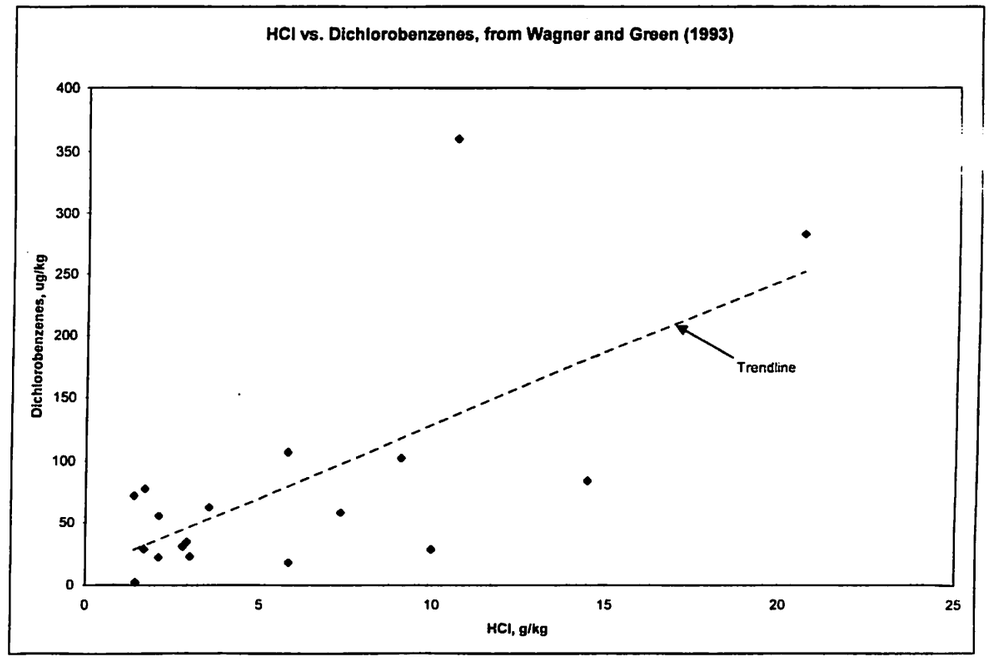

Figure 5: Experimental Data from Wagner and Green (1993)

3.5 Lenoir, D., Kaune, A., Hutzinger, O., Mutzenich, G., and Horch, K. 1991. Influence of operating parameters and fuel type on PCDD/PCDF emissions from fluidized bed incinerator. Chemosphere 23: 1491-1500.

Another of the seven studies said to support a threshold hypothesis, this study was characterized by Wikstrom et al. (1996) as follows:[46]

> *"No differences in emissions of PCDDs/PCDFs were found between the combustion experiments with or without NaCl in the fuel. However, when 3% PVC was added, the emissions of PCDDs/PCDFs were increased by a factor of 3. Other combustion experiments with three fuels that contain 0.1, 0.2, and 0.9% of chlorine were done, and all experiments contain approximately the same levels of PCDDs/PCDFs."*

Other than erroneously describing the lowest chlorine content as 0.1 percent rather than the 0.01 percent reported in the original paper, the above description is an accurate, albeit incomplete reflection of the findings by Lenoir et al. (1991). These scientists described their findings as follows:[47]

> *"The investigated fuel types varied in the chlorine content which, in some experiments, was increased by adding NaCl or polyvinylchloride (PVC). Only the addition of 3 % PVC to polyethylene resulted in an increase in PCDD/F concentrations. ... [A]ddition of 3 % PVC to PE increased PCDD/F levels by a factor of 3 compared to the mean value for pure PE combustion."*

Lenoir et al. (1991) added sodium chloride and/or PVC to cellulose fiber and polyethylene, extending the range of chlorine levels in these materials from 0.2 and 0.01 percent, respectively, to as much as 1.69 percent. According to Lenoir et al. (1991), these fuels, both modified and unmodified, were combusted under *"operating conditions [that] varied extremely."* Moreover, determinations were made for dioxin in flyash only, which, in two experiments, was found to vary *"between 25 and 81%"* of total dioxin in the flue gas. As a result, Lenoir et al. (1993) cautioned as follows:[48]

> *"[S]tatements about the influence of incinerator operating conditions and fuel types on PCDD/F emission might have to be modified if the total PCDD/F concentration in the flue gas, rather than the PCDD/F fraction attributed to coarse and fine fly ash, had been considered."*

Due perhaps to the extreme variations in operating conditions and only partial dioxin analysis, experimental results from this study are difficult to interpret. For example, the lack of correlation between percent chlorine in feed* and HCl emissions, as shown in Figure 6, is difficult to explain.

* Values for percent chlorine in feed were calculated from the data presented in Lenoir et al. (1991).

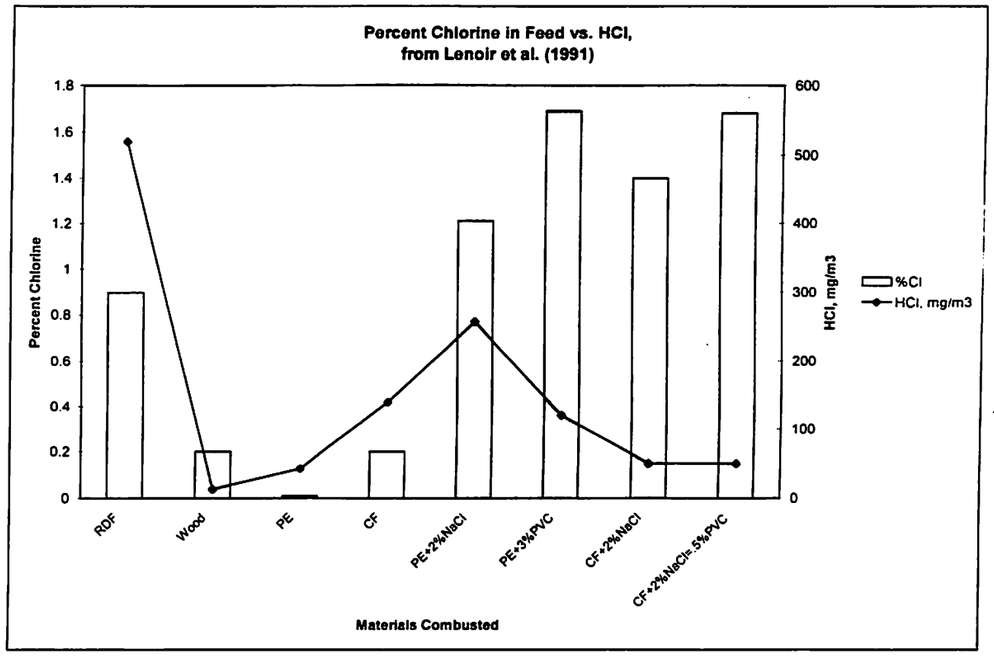

Figure 6: Experimental Data from Lenoir et al. (1991)

In summary, although the results obtained by Lenoir et al. (1991) are of great interest, it is difficult to see how they can be interpreted as indicating a threshold at 1 percent chlorine, as Figure 7 illustrates.

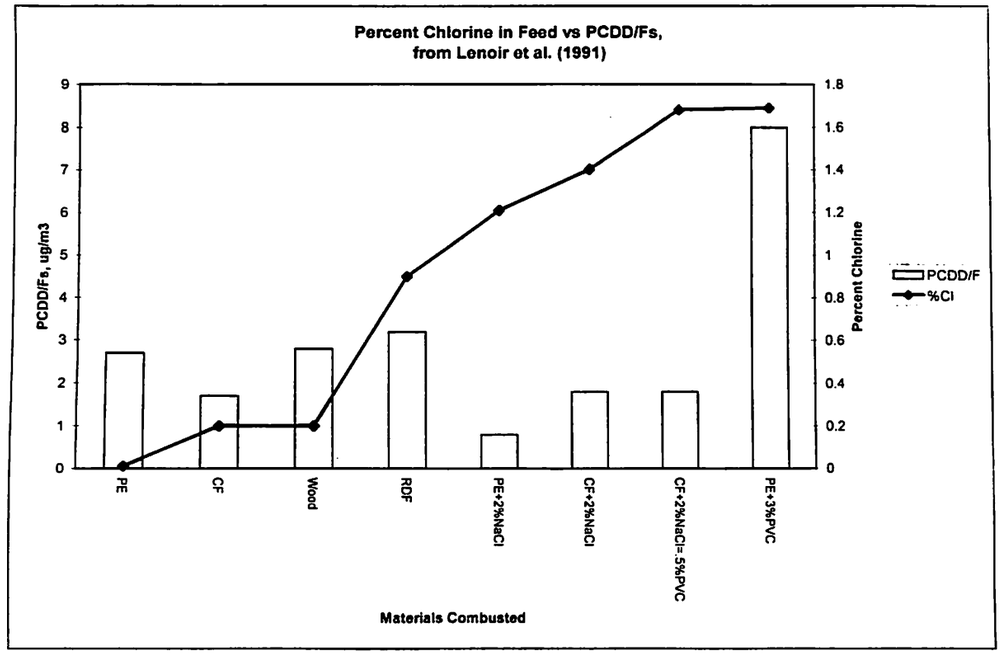

Figure 7: Experimental Data from Lenoir et al. (1991)

3.6 Ruuskanen, J., Vartiainen, T., Koja, I., Manninen, H., Oksanen, J., and Frankenhaeuser, M. 1994. Formation of polychlorinated dibenzo-p-dioxins and dibenzofurans in co-combustion of mixed plastics with coal: Exploratory principal component analysis. Chemosphere 28 (11): 1989-1999.

3.7 Frankenhaeuser, M., Manninen, H., Kojo, I., Ruuskanen, J., Vartiainen, T., Vesterinen, T.R., Virkki, J. 1993. Organic emissions from co-combustion of mixed plastics with coal in a bubbling fluidized bed boiler. Chemosphere 27 (1-3): 309-316.

These two studies, which relied upon the same experimental data, were also said by Wikstrom et al. (1996) to support their theory of a 1 percent threshold. The Swedish group described these studies as follows:[49]

"Ruuskanen et al. (27) and Frankenhaeuser et al. (28) studied the emissions of PCDDs/PCDFs during co-combust of coal with mixed plastic material. The plastics were added in five levels that correspond to 0, 0.6, 2.0, 2.7, and 3.9% of chlorine. No correlation between the chlorine level in the fuel and PCDDs/PCDFs was found when the chlorine contents in the fuels were 2.7% or lower (28). However, the emission level of PCDDs/PCDFs from the fuel with 3.9% chlorine was

about 10 times higher (27). These results indicate that the amount of chlorine in the fuel is of importance for the formation of PCDDs/PCDFs."

A careful examination of each of these studies failed to corroborate the values given above as the chlorine contents of the materials combusted in these experiments. According to data presented in Table 2 of the paper by Frankenhaeuser et al. (1993), only four levels of chlorine in feed were combusted and these levels were approximately 0.1, 0.5, 1.4, and 2 percent.

Frankenhaeuser et al. (1993) described their study and findings as follows:

"The objectives of this study were to examine the extent to which mixed plastics (4 % Cl) can be burned in a modern boiler together with coal (0.5% S), and to check the influence of the sulphur/chlorine ratio in the fuel on emissions. ... No clear correlation to increased mixed plastics feed could be seen [in flue gas emissions of PCDD/F]. PCDD/F in fly ash increased significantly with increased mixed plastics feed ... It is probable that the sulphur contained in the primary fuel poisoned the catalysts responsible for the formation of PCDD/F in all the plastics/coal blends."

Relying on the same data, Ruuskanen et al. (1994) represented their work and findings as follows:

"The purpose of the tests was to examine the effects of sulphur-rich coal on the formation of chlorinated hydrocarbons like PCDD/PCDFs. ...No correlation between HCl and furans and dioxins [in flue gas] was found. On the other hand, in fly ash samples the significant correlation of HCl and furans was obvious."

In summary, these studies found no correlation between chlorine input, as estimated by HCl emissions, and dioxin emissions. However, they did find a positive correlation between chlorine input and dioxin concentrations in fly ash. Consequently, while they cannot be said to support the theory of a threshold in the relationship between chlorine input and dioxin emissions, they do appear to support materials policies as effective measures for dioxin abatement.

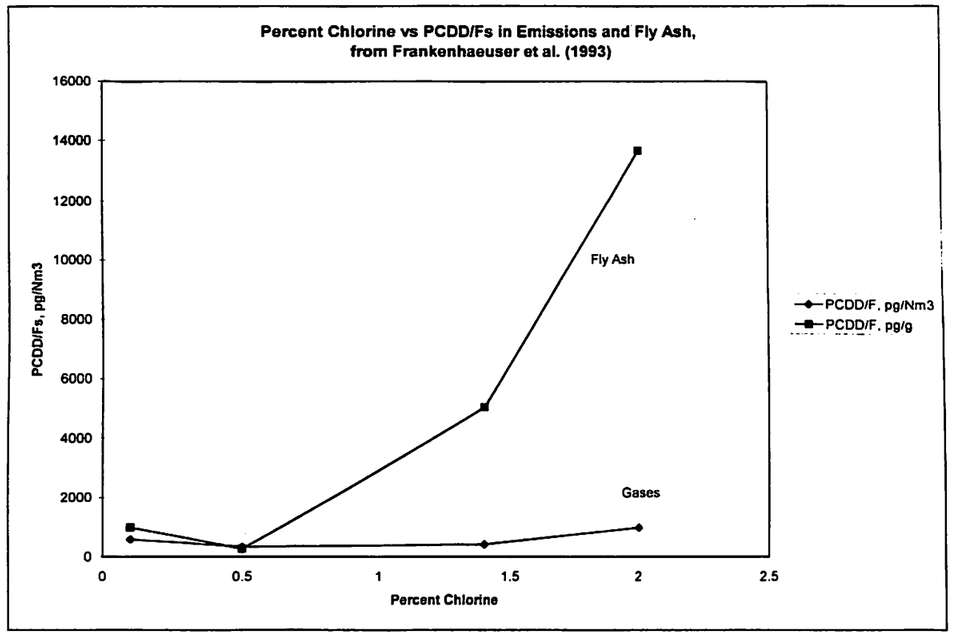

Figure 8: Experimental Data from Frankenhaeuser et al. (1993)

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Greenpeace International
Keizersgracht 176
1016 DW Amsterdam

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