Appendix 3: Full-Scale Combustors: Studies Relevant to the Relationship of Chlorine Input and Dioxin Output[a]

Hunsinger et al. (2000)[1]	*"Further experiments burning different fuels showed that inside the combustion chamber the formation of PCDD/F depends significantly on the fuel composition. Especially the content of halogens (and metals) play an important role. Burning bio mass fractions like natural wood chips or straw pellets resulted only in trace amounts of PCDD/F above the fuel bed. In contradiction to the measurements inside the combustion chamber the concentrations of PCDD/F found in the raw gas downstream of the boiler showed no influence of the fuel composition. The explanation of these results must be attributed to memory effects from fly ash deposits in the boiler section. They influence significantly the PCDD/F formation inside the boiler over a very long time."*
Yamamura et al. (1999)[2]	"*The furnace H emitted the lowest dioxin of 0.012ng-TEQ/m3N. This is due to not only the property of the waste, which are chemical residues with high volatile content, but also the characteristics of the furnace, which is easy to have stable combustion condition. Compared with E with the highest dioxin of 160ng-TEQ/m^3N and F, although F emits 2.5 times higher CO than E, F emits much lower dioxins of 2.9ng-TEQ/m^3N than E. One of the main reasons for this significant difference is due to the wastes incinerated. E incinerated plastics including medical waste, which contains plastic bags and tubes mainly made of PVC, and wood chips, while F incinerated demolition waste such as wood chips."*
Costner (1998)[3]	Findings in this evaluation of a hazardous waste incinerator (rotary kiln) in the U.S. are as follows: *"Data … show that PCDD/F emissions are dependent on chlorine input at a chlorine concentration as low as 0.031 percent. In addition, mass balance data indicate that hazardous waste incinerators may be achieving destruction efficiencies that are orders of magnitude below those achieved by other destruction technologies. … PCDD/F concentrations in stack gases increased as the chlorine content rose from 0.031 to 0.10 percent. … HCl/Cl_2 emissions do not correlate well with chlorine feedrates as has been assumed by some assessments of full-scale incinerators, e.g., Rigo et al."*
Manninen et al. (1997)[4]	*"A one- year co-combustion of RDF with peat and coal was carried out in a 65 MW circulating fluidized-bed (CFB) boiler power plant…. HCl emissions increased when the chlorine*

[a] This compilation of studies is not a fully comprehensive collection of all such studies on this topic.

	content of the fuel mixture increased, because limestone injection was not used."
Manninen et al. (1996)[5]	Results of seventeen test runs in a circulating fluidized bed boiler are as follows: *"Because a mixture of packaging materials can contain metals from foils and printing inks, chlorine from PVC, and food contaminants, a question has emerged: How do these metals and chlorine affect PCDD/F formation? On the other hand, does the combustion of packaging material with sulphur containing coal have an inhibiting effect on the formation? The objective of this study was to find answers to these questions by evaluating the full-scale combustion results from 17 test runs carried out in a multifuel circulating fluidized-bed boiler (CFB). Multivariate projection methods such as Principal Component Analysis (PCA) and Partial Least Squares Projections to Latent Structures (PLS) were used. … The results of the PLS calculations (-> Fig. 2 and regression coefficients) show that a lower heating value (LHV in dry solids) or the amount of volatile components in the fuel mixture, or the temperature of the bed did not correlate significantly with the PCDD/F values. Pb, Cu, Cr and Cl contents of the fuel and polychlorinated phenols (PCPhs) in the flue gas correlated most closely with the PCDFs in the flue gas and fly ash. … The chlorine content of the fuel correlated with PCDFs and there was an inverse correlation between the S/Cl ratio and PCDFs."*
Wanke and Vehlow (1996)[6]	This study at the TAMARA facility found that *"an additional input of Br [bromine] into the incinerator does obviously promote the specific formation of mixed halogenated [dioxin] congeners significantly. In Fig. 2 the sum of the Br containing dioxins [in raw gas] is plotted as a function of the Br input into the incinerator for all test trials except of those with poor combustion conditions. As a first approach the graph can be interpreted as a straight correlation between both parameters. The data, however, are very low and suffer from high analytical uncertainties."*
Huotari and Vesterinen (1996)[7]	In experiments performed with three commercial fluidized bed combustors and one lab-scale FBC, Huotari and Vesterinen (1996) found: *"The chlorine content of fuel rose up to 0.35% as a measured dry base (db). The emissions of the toxic PCDD/F compounds were found to follow fuel-Cl content trendwise, as expected. However quantitatively the relative emission levels varied with the factor of 10^3 at the same Cl- content in fuel. ... The PCDD/F-emissions are related to fuel-Cl content. … The PCDD/F-concentrations in flue gas (ng/m^3_n) tended to follow fuel Cl-content, but PCDD/F contents in separated ash (ng/g)*

	was mainly influenced by combustion conditions, like the furnace temperature. … The increase in concentrations of chlorinated benzenes, chlorophenols and PCB in flue gas, fuel-Cl and fuel-Cl/S and Cu in fly ash increased PCDD/F contents in flue gas. "
Vesterinen and Flyktman (1996)[8]	*"Refuse derived fuel (RDF) has been burned with wood chips and milled peat in a 4 MW bubbling fluidized bed boiler. … In co-combustion of RDF [refuse derived fuel] with wood chips the clear correlation was seen between the chlorine content of the fuel mixture and the PCDD/F concentration in flue gas. A similar correlation was also seen between the fuel chlorine and chlorophenol concentration in flue gas."* [Note: the chlorine content of the fuels to ranged from 0.03 to 0. 33%]
Moller et al. (1995)[9]	In their review of data from some of the 33 full-scale combustors in Denmark, Moller et al. (1995) concluded as follows: *"The hydrochloric acid content in the stack fume is dependent on the content of PVC and other chlorine sources in the waste. It is calculated that 2/3 of the chlorine in the Danish incinerator plants originates from PVC in the waste and thus also the hydrogen chloride formed. Burning PVC also yields a large amount of soot-containing smoke. ... The presence of chlorine in the material* [PVC] *gives the potential for contribution to the formation of polychlorinated dioxins and furans during the fire. If formed, it seems that these substances have the highest tendency to be present in the soot. The amount will depend on the fire conditions such as oxygen available, temperature, catalyst available such as copper and the amount of chlorinated material, e.g., PVC involved in the fire. However, the content of PVC in the waste also contributes to the emission of dioxins and heavy metals. … There seems to be a relationship between HCl emissions and the emissions of a number of chlorinated organic compounds including dioxins. However, the relationship is a 2nd or 3rd order relationship… . It is most likely that the reduction of the chlorine content in the waste can contribute to the reduction of the dioxin formation, even though the actual mechanism is not fully understood. The influence on the reduction is also expected to be a 2nd or 3rd order relationship."*
Thomas and Spiro (1995)[10]	In their study, Thomas and Spiro (1995) reviewed data from more than 80 full-scale combustion systems/processes in the U.S., concluding as follows: *"Nevertheless, the data presented in section 4 will show that dioxin emission factors from combustion of similar fuels, with broadly similar combustion*

	systems and pollution control devices, typically vary by less than an order of magnitude. This holds true, for example, for hospital waste incinerators, sewage sludge incinerators, and industrial wood boilers. Thus, for most processes dioxin emissions can be categorized by the combustion activity. Exceptions are the incineration of municipal and hazardous waste, for which different types of combustion systems can have dioxin emission factors that differ by more than an order of magnitude." "*While a dependence on chlorine content might seem to be a foregone conclusion, there has in fact been doubt on this point since limited data, on test-burns at municipal waste incinerators, for example, have shown no correlation. There are, of course, many variables besides chlorine content, especially the operating conditions of the combustion system, and the application of pollution control technology. Figure 1 shows that favorable operating conditions (open symbols) can lower emissions by as much as two orders of magnitude. But under poorly controlled conditions (closed symbols), there is a clear dependence on chlorine content. Moreover, the relationship is more than proportional since the slope of the loglog plot is somewhere between one and two. Since dioxin molecules contain more than one chlorine atom, a higher order dependence is not unexpected."*
Rigo et al. (1995)[11]	This study is described as examining data from a total of 169 full-, lab-, bench- and pilot-scale facilities. *"A general review … found no statistically significant relationship between chlorine input and PCDD/F stack gas concentrations for the majority (80 percent) of the 90 facilities which had sufficient simultaneous data … Eleven percent displayed an increase; 9 percent displayed decreasing PCDD/F concentration with increasing chlorine."* [Note: The lack of agreement between the conclusions presented in this report and the results of the authors' statistical analyses as well as many other limitations are addressed in detail in Costner (1997)[12] and, specifically with respect to municipal and medical waste incinerators, in Costner (1997).[13] As documented in these reviews, statistical analyses by Rigo et al. (1995) actually show a positive association between HCl and dioxin concentrations in stack gases at 15 of 22 municipal waste incinerators and 10 of 15 medical waste incinerators as well as a positive association between chlorine federates and dioxin concentrations in stack gases at 14 of 23 cement kilns.]

Cains and Dyke (1994)[14]	An examination of data from more than a dozen combustion facilities in the U.K. led to the following conclusions: *"Simple global correlations of dioxin and furan levels against general parameters such as the CO or HCl concentration in the flue gas, Cu, Cl or S levels in the feedstocks, grit ashes or particulates entrained in the flue gas, combustion or boiler exit temperatures are not sufficiently robust to characterize the conditions that promote formation in a general or universal way. Specific correlations of PCDD/F levels against the copper and chlorine contents of entrained particulates tentatively support results of laboratory studies on the role of Cu and Cl in fly ash in promoting PCDD/F formation."*
Mark (1994)[15]	This study was commissioned and published by the Association of Plastics Manufacturers in Europe and carried out at the TAMARA mass burn incinerator in Germany. *"In order to categorise the relative effects of polymers on the combustion of MSW the project studied three test cases:* • *combustion of MSW containing an average level of polymers - base case* • *combustion of MSW containing an added 7.5 per cent by weight of mixed plastics waste - medium polymer case* • *combustion of MSW containing an added 15 per cent by weight of mixed plastics waste - high polymer case."* *… APME estimates the European average content of PVC in MSW to be approximately 10 per cent weight, so to match this figure the PVC element of mixed waste polymers used for testing was increased by the addition of post-use articles such as PVC pipes and flooring products. This meant that the addition of plastics, with an average heat of combustion three times higher than standard MSW, caused the total feed to the combustor to be reduced accordingly.* *... soot blowing was carried out two hours before, and at the end of every six hour sampling period. Change over times from one condition to another - ie A to C - were a minimum period of 12 hours to ensure stable boiler conditions. … Two types of additives - lime, with or without carbon - were used during the test to confirm the effectiveness of carbon in removing dioxins and furans. … HCl concentration in the flue gas rose as expected because of the addition of PVC to the waste plastics, although this was subsequently reduced to amounts found in normal clean gas by the emission control system. … Peaks in raw gas concentrations were only two to three times the normal value of 1.5 g HCl/m^3. … There was no increased formation of chlorinated dibenzodioxin/furans (PCDD/F) in the raw gas when adding mixed waste plastics."*

	[Note: The data presented are not sufficient for assessing the relationship between chlorine and/or PVC input and dioxin output: 1) neither the chlorine content nor the actual PVC content of the incinerator feedstock was reported; 2) waste feedrates were apparently decreased as plastics content was increased; 3) additives were used -- lime with and without carbon -- that are commonly used to reduce dioxin formation by reacting with the available HCl and Cl_2 and/or reduce dioxin levels in the flue gas through adsorption; and 4) the frequency of soot blowing combined with abbreviated stabilization times (as little as 12 hours) can be expected to have considerable influence on dioxin formation as well as measured dioxin values.]
Wilken et al. (1994)[16]	At the Bielefeld-Herford municipal solid waste incinerator in Germany, reductions in PCDD/F emissions were achieved as follows: *" ... [T]he average of 2.73 ng I-TEQ/m^3 and the median of 2.81 ng I-TEQ/Nm^3 of the actual routine performance lie distinctly below former emission concentrations, which ranged from 2.9 to 4.2 ng I-TEQ/Nm^3. The same trend can be observed for PCB, but not for chlorobenzenes and chlorophenols … Since the measurement in December 1991 effective emission reduction means like* • *exclusion of PVC and computer scrap in the input,* • *optimizing combustion operation,* • *installation of a high-performance quasi-dry flue gas purification system in 1993 have been realized at this plant."*
Jager et al. (1993)[17]	In experiments at three full-scale combustors in Germany, Jager et al. found as follows: *"At MWI A, addition of polyvinylchloride resulted in a slight but not significant increase in PCDD/PCDF in clean gas, while concentrations of chlorobenzenes, PCB and hydrogen chloride increased rapidly. Nevertheless, the chlorine content in the input must have had an influence on the dioxin emission, as has been observed at MWI C at the summer and winter campaign. Lenoir et al. also observed a significant increase in PCDD/PCDF emission after addition of 3% PVC to the waste input. ... At MWIs A and D, the raise of the bromine content in the input material resulted in a significant rise of mixed-halogenated dioxins and furans, especially monobromo-polychloro compounds, and in increase of chlorinated dioxins and furans too (Fig. 5)."* *"Recommendations concerning waste input* • *Reduce halogen availability in the combustion chamber by excluding certain materials from MWI input.*

	• *Exclude brominated compounds (e.g., electronic scrap) from your MWI.* • *Exclude polyvinylchloride (PVC) from combustion in order to reduce halogen offer and PCB, chlorobenzene and hydrogen chloride emissions."*
Frankenhaeuser et al. (1993)[18]	This study was performed with a bubbling fluidized bed boiler. *"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."*
Thomas (1992)[19]	*"... [C]hlorine is not the only determinant of dioxin emissions. However, for larger variations in chlorine content, the data indicate a relationship between chlorine content and dioxin emissions. While there are sources with high chlorine content and low dioxin emissions, there are no known sources of low chlorine content and high dioxin emissions."*
Takeshita et al. (1992)[20]	At a full-scale municipal waste combustor, Takeshita et al. found as follows: *"The simultaneous control of combustion, hydrogen chloride (HCl) concentration level in the flue gas and the gas temperature proved very effective in the control of PCDD/Fs formation … [W]ith increasing gas temperature or HCl level in the gas except for exp. 4, the PCDDs/Fs concentrations tended to increase, and at any of similar gas temperature, the concentrations tended to increase at higher HCl levels. However, the PCDDs/Fs concentrations in the EP ash samples were closely related to the amount of injected slaked lime. ... At any of similar gas temperature, the amounts of PCDDs/Fs formed at the high HCl level were higher than those measured at the middle HCl level, and at similar middle and high levels of HCl, the amounts of PCDDs/Fs formed at higher gas temperatures were higher than those formed at lower gas temperatures. At a gas temperature of 300 ^{o}C, HCl in the flue gas markedly increased the formation of PCDDs/Fs. ... The PCDDs/Fs concentrations in the gas when the gas temperature was 300 ^{o}C were at higher levels in proportion to the HCl concentration levels. When the gas temperatures were 200 and 240 ^{o}C, the PCDDs/Fs concentrations at the lowest HCl level in the gas were remarkably higher than at*

	other HCl levels. This phenomenon indicates that the PCDDs/Fs formation was linked to some other factors increasing the amount of dust in the gas as shown in Fig. 8."
Johnke and Stelzner (1992)[21]	In their study of 15 full-scale municipal waste combustors in Germany, Johnke and Stelzner found as follows: *"The addition of PVC (1% wt) to the waste resulted in a marked increase in HCl levels in the raw gas, but in no significant change in PCDD/PCDF concentrations. However, it cannot be concluded from this that PCDD/PCDFs do not form in the incineration of PVC. ... Tests involving an increased supply of bromine in the waste have also been performed; here, a distinct increase in polyhalogenated dioxin/furan levels and a slight increase in the concentrations of the chlorinated PCDD/PCDFs as well as ben-zenes, was ascertained."*
Matilla et al. (1992)[22]	In their study using a full-scale combustor, Mattila et al. reported as follows: *"Our results prove the evidence for conversion of chlorinated plastic material to polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). ...[T]he total amount of PCDD/Fs correlates 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."*
Kopponen et al. (1992)[23]	At a 60 megawatts district power plant, Kopponen et al. (1992) combusted a mixture of coal and bark mixture with the addition of PVC and non-chlorinated plastics. *"... the total PCDD/PCDF concentrations were over 100 times higher in the fly ash from combustion of chlorine containing material (combustion 4) than those of basic fuel (coal and bark materials)."*
Manscher et al. (1990)[24]	*From their analysis of data from 10 municipal solid waste incinerators and two hospital incinerators in Denmark, Manscher et al. concluded as follows:* "For models that, in addition to the block effects, contain the parameters of load and excess air, it is found that among the rest of the parameters considered, only the hydrogen chloride becomes significant [for dioxin emissions]. ... [They found] ... a 14 % increase in the dioxin emissions when the HCl emission is doubled. This relation is valid in the range of 100 to 1000 mg/m^3 of HCl."

Vikelsoe et al. (1990)[25]	In experiments at a full-scale combustor, Vikelsoe et al. found as follows: *"The results showed increased dioxin emission at high levels of PVC and NaCl, and decreased emission at high lime levels."* [Note: When the PVC content of municipal solid waste was doubled, dioxin emissions (only PCDDs were measured) increased by 32 percent and HCl emissions by 64 percent. Doubling NaCl concentrations led to a 17 percent increase in dioxin emissions (PCDDs only) and a 9 percent increase in HCl. When HCl concentrations were doubled, dioxin emissions (PCDDs only) increased by 42 percent.]
Aittola and Wihersaari (1990)[26]	In this study, wood chips, wood chips with 65% crushed municipal waste, and wood chips with 85% crushed municipal waste were converted into gases in a 5 MW gasification unit and the gases were burned in a conventional boiler. The wood chips contained 0.3 percent by weight chlorine and the MSW, 12 percent by weight chlorine. Among the findings were *"the concentration of hydrochloric acid in the flue gases follows the mixing ratio of MSW."* Similarly, dioxin concentrations in the flue gases also increased with rising MSW content in the materials burned.
National Environmental Protection Agency, Denmark, (1989)[27]	In their study of full-scale combustors, the Danish EPA noted a 14 percent increase in dioxin emissions from municipal waste combustors and a 25 percent increase from medical waste incinerators when HCl concentration doubled.
Takeshita and Akimoto (1989)[28]	A study with a fluidized bed incinerator in Japan found as follows: *"PCDDs and PCDFs in the boiler and the EP [electrostatic precipitator] ash samples and in the gas samples were found at lower levels in proportion to the decrease of levels of hydrogen chloride in the gas. ... [L]evels 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.... From the relationship between hydrogen chloride levels in the gases at the EP outlet and the total amounts of PCDDs and PCDFs as shown in Figure 11, it is clear that the control of hydrogen chloride levels by the supply of Dolomite to the furnace controlled the formation of PCDDs and PCDFs successfully."*
Giugliano et al. (1989)[29]	Tests in a *"non-conventional system",* an industrial rotary kiln with an afterburner led to the following conclusions: *"The incineration of urban wastes with added PVC, up to 10% by*

	weight, did not produce the emission of detectable concentrations of polychlorinated dibenzodioxins and -furans upstream flue gas cleaning equipment. ... The tests were conducted in a 1.5 t h^{-1} industrial rotary kiln/afterburner combustion chamber, equipped with a fuel oil burner of high heat output that makes the unit a nonconventional system particularly interesting for the thermal destruction of organic micropollutants. ... Sampling was performed upstream the air pollution control equipment ... For organic micropollutants, 3.5 to 5 m^3 of gas at normal conditions were collected in duct with an allglass isokinetic probe followed by a glass wool filter for particulate matter retention, two consecutive water cooled flasks at 2 ^oC and a final C-18 adsorbent cartridge. ... Detection limits of the analytical procedure employed are 0.5 ng for 4- to 7-CDDs and -CDFs, 2.5 ng for 8-CDDs and -CDFs, ..." [Note: The failure to detect any PCDDs can be attributed to reliance on an analytical procedure which, by today's standards, had unusually high detection limits, as shown above. Also PCDFs were not analyzed.]
Gottesman et al. (1988)[30]	This study, entitled "Vinyl Industry Response to Environmental Concerns About PVC in Municipal Solid Waste," had the following findings: *"As expected, the amount of PVC in the waste being incinerated does increase the amount of HCl in the combustion gases. ... In this, and the Prince Edward Island Study, the data show that the amount of dioxin that finally came out of the incinerator in these test runs was approximately the same amount as that that came in with the MSW. In other words, there was no net increase in environmental dioxins as a result of the incineration process. ... At the tertiary duct and the boiler outlet, none of the contrasts associated with the amount of PVC were significant at the 5% level for either PCDDs and PCDFs."* [Note: This study has been cited by some authors as having found no correlation between chlorine input and dioxin output. Since the authors do not present actual input or output data, their findings cannot be reviewed. However, their last statement shown above leaves open the possibility that a positive correlation was found between the amount of PVC combusted and dioxin concentrations at the tertiary duct and boiler outlet but that this positive correlation was significant at some level below 95 percent.]
Karasek and Viau (1983)[31]	This study was carried out with an energy recovering incinerator in France. ***"****Data shown here are the result of addition of 300% the usual level of PVC to a municipal*

	incinerator. No significant differences in the concentrations of these substances [PCDDs] *on the fly ash were found.... it is evident that in these experiments the addition of PVC to the feedstock had no observable effect on the levels of PCDD, and other compounds generated in the incineration process. … Unspiked garbage was found to contain 1% PVC of the total feed by laboratory analysis. … It is striking that there is a distinctive lack of hydrocarbons and polycyclic aromatic hydrocarbons … "* [Note: The analytical method used was, as expected, primitive with high detection limits. Also PCDFs were not determined.]
U.S. Environmental Protection Agency (1984)[32]	*"The chlorine content of fuel is obviously an important parameter affecting the formation of PCDD's and PCDF's. However, it can be generally stated that chlorine must be present for the formation of PCDD, and general trends indicate that increased chlorine concentrations in the feed improve the possibilities of PCDD emissions. … Shih et al. developed a ranked priority list of conventional combustion systems emitting polycyclic organic matter including PCDDs and PCDFs. The rationale presented for source ranking is based on fuel characteristics and combustion conditions. Shih's work places great emphasis on both the chlorine content of the feed and the concentration of aromatics in the feed."*
Olie et al. (1977)[33]	*"During our investigation of the environmental loading with organic pollutants by waste products of municipal incinerators we detected a large number of organochlorine compounds in fly ash ... [The most abundant chlorinated compounds in all fly ash samples were highly chlorinated benzenes ... The amount of chlorodibenzodioxins and chlorodibenzofurans entering the atmosphere via flue gases is probably quite small. However, due to the extreme toxicity of some of the components and the fact that most incineration plants are located in densely populated areas extensive monitoring of such facilities might be advisable."* [Note: Table 1 of this report shows all tetra- through octa-PCDDs and PCDFs were detected in the fly ash of the Arnheim incinerator, while Table 2 shows that all tetra- through octa-PCDDs were detected in the flue gas of the Alkmaar incinerator.]

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