

Prevention of Polychlorinated Dibenzop-dioxins/Dibenzofurans Formation on Municipal Waste Incinerator Fly Ash Using Nitrogen and Sulfur Compounds

RUUD ADDINK,*
RUDI H. W. L. PAULUS, AND
KEES OLIE

*Department of Environmental and Toxicological Chemistry,
Amsterdam Research Institute for Substances in Ecosystems,
University of Amsterdam, Nieuwe Achtergracht 166,
1018 WV Amsterdam, The Netherlands*

Formation of polychlorinated dibenzo-*p*-dioxins (PCDD) and polychlorinated dibenzofurans on incinerator fly ash can be reduced under laboratory conditions by the addition of ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA), or Na₂S. With these compounds (inhibitors), a reduction in formation of ca. 80–90% is achieved. The inhibitors reduce the ratio of [PCDD]:[PCDF] formed, suggesting that more than one pathway for the formation from carbon exists. EDTA and Na₂S reduce the degree of chlorination of PCDD; for PCDF, the same effect is seen with Na₂S. No change was ever seen in the isomer distribution within homologues as a result of the inhibitors added, in accordance with the theory that these isomer distributions are thermodynamically controlled. In additional experiments, NTA gave a good reduction with reaction times of 30–60 min, at temperatures between 300 and 400 °C, and with concentrations in the reaction mixture of 2–10%. With HCl as a chlorinating agent, the inhibition by NTA is >50% for both PCDD and PCDF. These inhibitors show promising results to achieve the reduction of PCDD/F formation. The additional experiments with NTA show that this compound can reduce formation under various conditions, making it suitable for use in the post-combustion zone of an incinerator, where reaction conditions will vary.

Introduction

In 1977, polychlorinated dibenzo-*p*-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) were detected in emissions of waste incineration plants (1). Theoretical calculations showed that these toxic compounds are formed

through a heterogeneous mechanism at lower temperatures (<600 °C) rather than via homogeneous gas phase reactions (2, 3). Such a temperature regime exists in the post-combustion zone of the incinerator. The off-gas from the combustion chamber passes the part of an incineration plant in which these gases are cooled down and passed through an air pollution control device before being emitted from the stack.

Residue particles (fly ash) present in the off-gas act as an active surface for the formation reactions of PCDD/F. Laboratory studies suggest that transition metal ions like Cu and Fe catalyze formation reactions, which occur on the fly ash particles (4). Formation can take place from both macromolecular carbon already present on the fly ash surface (4) and small organic molecules (precursors) like chlorophenols (5). These precursors can be adsorbed onto the fly ash from the off-gas and subsequently converted to PCDD/F. Chlorine incorporated into the DD/F structure appears to originate both from metal chlorides present in fly ash (6) and from gases like HCl (7).

Prevention of PCDD/F formation during the incineration process is called inhibition. Compounds capable of inhibition are called inhibitors. From a technical viewpoint, the concept of inhibition differs from flue gas cleaning techniques in that inhibition deals with prevention of PCDD/F formation, whereas flue gas cleaning removes PCDD/F already formed.

Two categories of inhibitors have been developed and tested under laboratory conditions. The first category is formed by basic compounds and includes NH₃ (8), CaO (9), KOH, and Na₂CO₃ (10). The mechanism involved might be that these inhibitors change the acidity of the fly ash surface. Milligan found that PCDD/F formation decreases as the fly ash is more alkaline (11). Compounds that are likely to form some kind of complex with the transition metal ions that catalyze PCDD/F formation are a second type of inhibitor. One approach is to use sulfur compounds for this purpose. Both CS₂ and thiophene are capable of reducing PCDD formation from pentachlorophenol on fly ash (12). Functionalized amines, e.g., ethanolamine and triethanolamine, are good inhibitors as well. These compounds reduce PCDD formation from pentachlorophenol on fly ash to less than 1% (13). Functionalized amines are also capable of reducing PCDD/F formation from pentachlorophenol on industrial incinerator fly ash (14). Other papers have reported on the inhibition of PCDD/F formation without disclosing the nature of the compounds used (15, 16). An interaction (complexation) with the (Cu) catalyst is the most likely manner in which the functionalized amines work (17).

It should be pointed out that all the studies mentioned above were carried out in laboratories, typically by heating fly ash samples, mixed with reactants and inhibitors, in a glass tube under a gas stream containing nitrogen and oxygen. The use of inhibitors in actual incineration plants appears to be limited so far. Some studies have reported on use of inhibitors, which were injected into the off-gas in the post-combustion zone (18, 19), resulting in reductions of >80% for both fly ash and off-gas. The nature of the inhibitor has not been disclosed.

* Corresponding author present address: Isermann Department of Chemical Engineering, Rensselaer Polytechnic Institute, Troy, NY 12180; telephone: (518) 276-6377; fax: (518) 276-4030.

TABLE 1

Testing of Various Inhibitors^a

exp no. ^b	inhibitor	ΣPCDD	ΣPCDF	[PCDD]:[PCDF] ^c	% PCDD inhibition ^d	% PCDF inhibition ^d
1	none	0.61 ± 0.06	4.96 ± 0.50	0.12		
2	EDTA	0.09 ± 0.01	1.27 ± 0.13	0.07	84 ± 3	77 ± 2
3	NTA	0.05	1.07	0.04	92 ± 1	78 ± 2
4	Na ₂ S	0.09 ± 0.01	1.01 ± 0.03	0.09	85 ± 2	79 ± 3
5	Na ₂ S ₂ O ₃	0.50 ± 0.02	3.37 ± 0.31	0.15 ± 0.01	17 ± 11	31 ± 13

^a Concentrations in nmol/g of fly ash. ^b All experiments: $T = 348 \pm 7$ °C; 60 min; 2.0 g of 93% fly ash/2% C/5% NaCl or 91% fly ash/2% C/5% NaCl/2% inhibitor; flow N₂ 53 ± 3 mL/min, O₂ 6.5 ± 0.7 mL/min, H₂O(g, not quantified); all experiments in duplicate, mean value ± range is shown; no range is given for ΣPCDD and ΣPCDF if negligible (<0.005). ^c Calculated for each run. This yields two values for each duplicate experiment. Mean value ± range is given, except if range is negligible (<0.005). ^d Relative to experiment 1 (no inhibitor), e.g., 25% formation (ΣPCDD or ΣPCDF) left is 75% inhibition.

TABLE 2

Testing of NTA under Various Conditions

exp no. ^a	parameter varied	ΣPCDD ^b	ΣPCDF ^b	% PCDD inhibition ^c	% PCDF inhibition ^c
	reaction time				
6 ^d	30 min, no NTA	0.32 ± 0.04	0.32 ± 0.18		
7 ^d	30 min, NTA	0.02 ± 0.01	0.61 ± 0.35	92 ± 5	82 ± 11
1 ^d	60 min, no NTA	0.61 ± 0.06	4.96 ± 0.50		
3 ^d	60 min, NTA	0.05	1.07	92 ± 1	78 ± 2
	temperature				
8 ^e	298 °C, no NTA	0.17 ± 0.04	1.11 ± 0.14		
9 ^e	298 °C, NTA	0.01 ± 0.01	0.08	90 ± 8	92 ± 1
1 ^e	348 °C, no NTA	0.61 ± 0.06	4.96 ± 0.50		
3 ^e	348 °C, NTA	0.05	1.07	92 ± 1	78 ± 2
10 ^e	398 °C, no NTA	0.47 ± 0.14	3.48 ± 0.43		
11 ^e	398 °C, NTA	0.01	0.23 ± 0.09	97 ± 2	93 ± 4
	% inhibitor				
12 ^f	no NTA	0.11 ± 0.04	1.13 ± 0.43		
13 ^f	2% NTA	0.01	0.27 ± 0.02	94 ± 3	72 ± 12
14 ^f	5% NTA	0.01 ± 0.02	0.09 ± 0.07	83 ± 17	88 ± 11
15 ^f	10% NTA	0.001	0.06 ± 0.05	99 ± 1	92 ± 7
	chlorine source				
16 ^g	no NTA, HCl	0.05 ± 0.03	0.37 ± 0.09		
17 ^g	2% NTA, HCl	0.01	0.11 ± 0.02	72 ± 22	68 ± 14

^a Experiments 1, 3, and 6–11 were performed in duplicate and experiments 12–17 in triplicate. Mean value ± range (for duplicates) or mean value ± s (for triplicates) is given. No range or s is given for ΣPCDD or ΣPCDF when negligible (<0.005). ^b Concentrations in nmol/g of fly ash. ^c Relative to the experiments without inhibitor under identical conditions, e.g., 25% formation (ΣPCDD or ΣPCDF) left is 75% inhibition. ^d $T = 348 \pm 7$ °C; 30–60 min; 2.0 g of 91–93% fly ash/2% C/5% NaCl/0–2% inhibitor; flow N₂ 53 ± 3 mL/min, O₂ 6.5 ± 0.7 mL/min, H₂O(g, not quantified). ^e See footnote d, but with $T = 298$ – 398 °C; 60 min. ^f $T = 298 \pm 7$ °C; 60 min; 2.0 g of 84–93% fly ash/2% C/5% NaCl/0–10% inhibitor; flow N₂ 111 ± 4 mL/min, O₂ 11 ± 2 mL/min, H₂O(g, not quantified). ^g $T = 298 \pm 7$ °C; 50 min; 2.0 g of 96–98% fly ash/2% C/0–2% inhibitor; flow N₂ 111 ± 4 mL/min, O₂ 11 ± 2 mL/min, HCl 5.1 ± 0.6 mL/min.

In the present paper, we report on a series of experiments that were carried out with compounds to test their ability to reduce PCDD/F formation on fly ash. Since Cu appears to be the strongest catalyst for PCDD/F formation, at least for PCDD/F formation from carbon (4), we chose to study ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA), Na₂S, and Na₂S₂O₃. These compounds are known to form stable complexes with Cu ions (20). These four compounds were first tested under identical conditions, and the amount of PCDD/F formed in these experiments was compared with experiments where no inhibitor was present. Subsequently, a series was performed with one inhibitor under varying reaction conditions (concentration of the inhibitor, reaction time, and temperature). Preliminary results of this research have been published elsewhere (21, 22).

Experimental Section

Materials. The chemicals used in our cleanup have been described before (23). The following chemicals were used in our experiments: activated carbon (Darco G-60, 100–325 mesh, Aldrich Chemie, Steinheim, FRG); ethylenediaminetetraacetic acid (99.5%, Aldrich Chemie, Axel, The

Netherlands); fly ash from a municipal waste incinerator in Zaanstad, The Netherlands; hydrogen chloride (gas, 4.0 grade, UCAR, Nieuw-Vennep, The Netherlands); nitrilotriacetic acid (99%, Aldrich Chemie, Axel, The Netherlands); nitrogen (5.0 grade, Hoekloos, Schiedam, The Netherlands); oxygen (4.5 grade, Hoekloos); sodium chloride (99%, Aldrich-Chemie, Steinheim, FRG); sodium sulfide (99.5%, Aldrich Chemie, Axel, The Netherlands); sodium thiosulfate (99%, Aldrich Chemie, Axel, The Netherlands).

Experimental Apparatus. Experiments are reported in Tables 1 and 2. From the fly ash, all organic material was removed by heating at 550 °C for 90 min under a stream of air saturated with water. The fly ash was subsequently mixed with activated carbon and NaCl by shaking. Reaction mixtures consisted of 84–98% fly ash, 2% carbon, 5% NaCl, and 0–10% inhibitor. No NaCl was used in experiments 16 and 17 with HCl in the gas flow. A total of 2.0 g of the mixture was placed in a cylindrical sample basket and coupled with a glass inlet tube for introduction of a gas flow through the fly ash bed. Sample basket and inlet tube were fit into a horizontal Pyrex glass reactor and put in a furnace (Lenton CSC 1100 split tube furnace with PID 808 temperature controller, Leicestershire, U.K.). A gas stream

(N₂, O₂) was passed through a washing bottle containing water and passed over the fly ash bed. In experiments 16 and 17, a mixture of N₂, O₂, and HCl (no water) was used. The flow was controlled by Series 840 Side=Trak mass flow controllers (Sierra Instruments, Monterey, CA). The flow was checked before and after experiments with a flow meter. The gases were mixed in a mixing chamber (V = 800 mL) containing ceramic pellets. Experiments lasted for 30–60 min, preceded by 10 min of heating in order for the sample basket, inlet tube, and reactor to reach the set point temperature (298–398 °C, accuracy ± 7 °C). During these 10 min, the gas stream was already passing through the fly ash bed. Products evaporating from the fly ash surface were collected using a cold trap (80 mL of toluene cooled with ice). After the experiment, the fly ash bed was taken out of the furnace immediately and cooled to room temperature. The fly ash fractions were combined with the cold traps before the beginning of the Soxhlet extraction.

Cleanup and Analysis. These have been described before (23). Only the T₄CDD–OCDD and T₄CDF–OCDF congeners were analyzed. The cleanup procedure, fly ash, carbon, NaCl, and all inhibitors were checked for background PCDD/F. Results varied between 0.002 and 0.026 nmol/g fly ash of ΣPCDD/F.

Results and Discussion

Testing of Various Inhibitors. Since carbon is an important source of PCDD/F formation and optimum temperatures of 350 °C have been reported for this reaction pathway (24), we chose to test the four compounds using a mixture of activated carbon and fly ash heated at 348 °C. The residence times of collected fly ash particles in the post-combustion zone of an incinerator can vary between a few min and several hours (25); we conducted our experiments for 60 min. Water is always present during the process of waste incineration and therefore added to the gas stream passing over the fly ash bed.

The mechanism of formation of PCDD/F from carbon is not clear at this time. Various routes might exist: Jay et al. suggested direct release of PCDD/F from the carbon, implying that these structures already exist in carbon (26); Luijk found that PCDD can be formed from carbon through chlorophenols as intermediates (27). Some publications (28, 29) have suggested that PCDD/F formation from carbon can be an artifact, the actual formation resulting from precursor molecules that are adsorbed on the carbon surface. However, we found that various small organic compounds (chlorobenzenes, DF, hexane, toluene) were not capable of PCDD/F formation on our fly ash unless a gaseous chlorine source (HCl) is present. We tested these precursors under reaction conditions similar as used in our inhibition experiments, and no significant PCDD/F formation was ever seen without added HCl. Therefore, we believe that precursors possibly adsorbed on activated carbon are not capable of producing PCDD/F as long as no HCl is added to the gas stream and that PCDD/F formed stem from activated carbon rather than from adsorbed precursors. The results found in our investigation allow us to draw some conclusions regarding the various possible formation mechanisms with carbon as reactant (see below).

Table 1 shows the results of our experiments with EDTA, NTA, Na₂S, and Na₂S₂O₃ as inhibitors. The percentage of inhibition relative to uninhibited experiments is reported for both PCDD and PCDF. EDTA, NTA, and Na₂S are equally good inhibitors for both PCDD and PCDF with ca. 80–90%

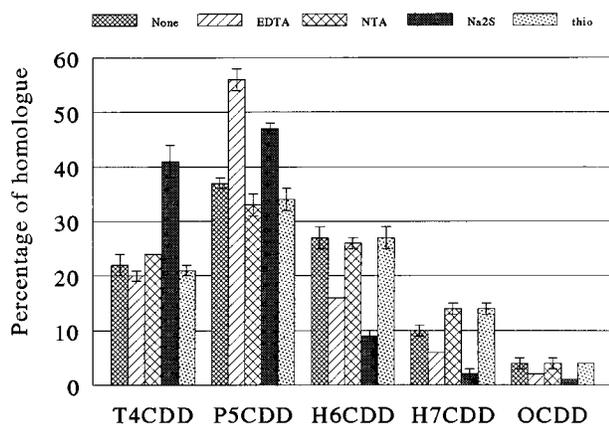


FIGURE 1. PCDD homologue distribution with various inhibitors (Σ PCDD = 100%, thio = Na₂S₂O₃).

reduction, whereas Na₂S₂O₃ is less efficient in diminishing PCDD/F formation. An explanation for these observations is not easy to give, as it is not clear whether these compounds remain intact on fly ash during the inhibition process. The role of the CH₂COOH groups in EDTA and NTA might be that a good adsorption or chemisorption is achieved, e.g., by H bonds. Dissociation of the two sulfur salts would yield S²⁻ and S₂O₃²⁻ and perhaps a difference in reactivity toward Cu.

As can be seen in Table 1, the [PCDD]:[PCDF] ratio decreases as a result of the addition of inhibitors, except for Na₂S₂O₃, which shows a slight increase. As described above, with PCDD/F formation from carbon, several pathways might exist, possibly with different catalysts. If an inhibitor shows better interaction (complexation) with one catalyst than with another, i.e., reduces PCDD/F formation through one pathway more than via another pathway, this will obviously effect the [PCDD]:[PCDF] ratio. Hence, the observation that this ratio changes as an effect of the inhibitor addition excludes PCDD/F formation via a single mechanism or catalyst.

An interaction with the catalyst could not only reduce the amount of PCDD/F formed but also reduce the degree of chlorination because less metal ions are available to catalyze chlorination. This is indeed observed with EDTA and Na₂S, which show a shift toward lower chlorinated homologues as compared with the uninhibited experiment for PCDD (Σ PCDD = 100%). With PCDF, the same effect is seen with Na₂S. With EDTA and NTA, there is an increase of T₄CDF (relative to the uninhibited experiment), but for the other PCDF homologues the differences are small. These results are depicted in Figures 1 and 2 (Σ PCDD or Σ PCDF = 100%). The addition of inhibitors may be viewed as being identical with decreasing the Cu concentration. With a decreasing Cu concentration, Stieglitz et al. also found a trend toward lower chlorinated DD/F, using a fly ash model mixture of Mg–Al–silicate, charcoal, Cu, and KCl (30).

In Figure 3, the influence of the four inhibitors on the isomer distribution within the H₆CDD homologue is presented. Note that not all isomers can be separated on the GC column. Σ H₆CDD was set to 100%, and the contribution of each isomer was calculated. This allows us to compare the isomer distributions found with the various inhibitors. As can be seen, the distribution does not depend on the presence of the inhibitor and is equal for all experiments. The same results were observed for the other homologues (not shown). Hence, the inhibitors appear to

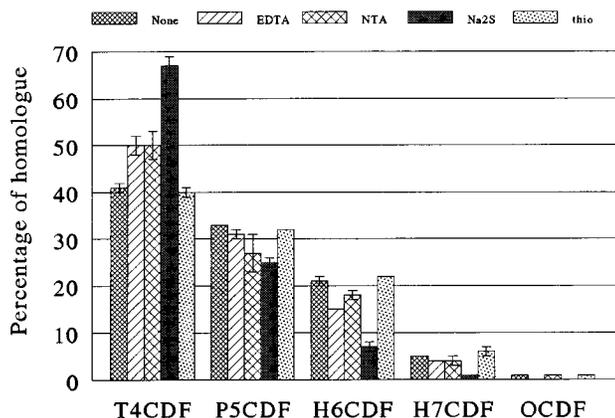


FIGURE 2. PCDF homologue distribution with various inhibitors (Σ PCDF = 100%, thio = $\text{Na}_2\text{S}_2\text{O}_3$).

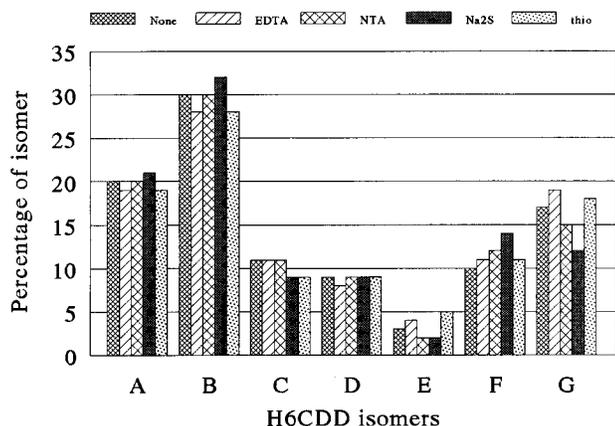


FIGURE 3. H_6CDD isomer distribution with various inhibitors. Explanation of letters: A = 1,2,4,6,7,9 + 1,2,4,6,8,9 + 1,2,3,4,6,8; B = 1,2,3,6,7,9 + 1,2,3,6,8,9; C = 1,2,3,4,7,8; D = 1,2,3,6,7,8; E = 1,2,3,4,6,9; F = 1,2,3,7,8,9; G = 1,2,3,4,6,7.

be capable of changing the [PCDD]:[PCDF] ratio and the homologue distribution but not of altering the isomer distribution within homologues. In other words, these compounds influence the amount of Cl built into the PCDD/F molecules [probably by reducing the number of Cu (or other metal) ions available for catalysis] but not the positions that are chlorinated. This is consistent with earlier observations that isomer distributions of PCDD/F formed on fly ash in general do not depend on the reaction conditions, especially are independent of the reaction time, and appear to be thermodynamically controlled (31).

Different Reaction Conditions with NTA. A second series of experiments was performed with one inhibitor (NTA) to study the influence of the reaction conditions on the degree of inhibition since in the post-combustion zone various reaction conditions may exist. Results are presented in Table 2. Experiments 6, 7, 1, and 3 show that the degree of inhibition does not change as a result of increased reaction times; loss of inhibitory activity, e.g., through dissociation of catalyst-inhibitor complexes, is not observed. Diffusion of the inhibitor through the fly ash bed could be limiting its activity if the time scale of the formation reactions of PCDD/F is short (seconds). However, formation from carbon is a slow process, which can continue for several hours, and diffusion of the inhibitor is not important.

Varying the temperature between 298 and 398 °C (experiments 8, 9, 1, 3, 10, and 11) has no strong effect on the degree of PCDD and PCDF inhibition. At the highest

temperature the NTA will evaporate from the fly ash surface more quickly. We studied the weight loss of a reaction mixture containing NTA by determining its weight before and after the experiment. The weight loss during the experiment increased from 39% at 200 °C, 74% at 300 °C, to 97% at 400 °C. The weight loss stems from desorption and/or decomposition. These results show that probably only a small fraction of the inhibitor added is active in reducing PCDD/F formation.

Concentrations of NTA were varied between 2 and 10% (experiments 12–15). Inhibition of PCDD formation is not higher with 10% than with 2% NTA. In both experiments, the percentage of inhibition is >90%. With PCDF, no conclusions can be drawn because of the margins of error. The results found for PCDD inhibition suggest that already with 2% NTA a surplus of inhibitor is present and increased concentrations have no effect.

In experiments 16 and 17, HCl was used as a chlorinating agent instead of NaCl, since HCl is always present in incinerator off-gas and is a strong chlorinating agent. Percentages of inhibition are >50%. These results show that NTA can effectively reduce chlorination by such a strong agent as HCl, which is an important chlorine source in flue gas. Note that with HCl present, precursors adsorbed on the activated carbon (as described above) might contribute to the PCDD/F formation. However, this does not affect the validity of the percentage of inhibition found in this experiment.

The conclusions drawn above regarding the [PCDD]:[PCDF] ratio, homologue distributions, and isomer distributions (based on experiment 3, compared with the uninhibited experiment 1) are valid for all the experiments reported in Table 2 also. For all these experiments, we observed a decrease of the [PCDD]:[PCDF] ratio as a result of added NTA, no change in the PCDD homologue distribution, a small shift toward lower chlorinated congeners for PCDF, and no change in any isomer distribution (not shown). Hence, variation of the time, temperature, inhibitor concentration, or chlorine source did not effect these results.

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