MORE THAN 10 YEARS CONTINUOUS EMISSION MONITORING OF DIOXINS BY LONG-TERM SAMPLING IN BELGIUM AND EUROPE - EXPERIENCES, TRENDS AND NEW RESULTS

Reinmann J

Environnement S.A Deutschland, Benzstrasse 11, 61352 Bad Homburg, Germany

Introduction

In the years 1999 and 2000 the governments of Flanders and Wallonia decided to implement by law the continuous emission monitoring of dioxins and furans for all domestic waste incinerators. In Flanders this demand was defined in Vlarem II[1] Art. 5.2.3.3.6 and in Wallonia according a decree dated on 11th May 2000[2]. Later this legal demand was extended to hazardous waste incinerators, cement kilns and other facilities. At this time Belgium was the only country in the world with a legal requirement for continuous dioxin emission monitoring and up to now only in Belgium more than 50 systems were installed in the different facilities.
In the meantime France followed the example of Belgium. A decree dated 3rd August 2010[3] demands by law the continuous PCDD/PCDF emission in all domestic and hazardous waste incineration plants until the 1st July 2014. This law covers around 200 stacks, which will increase the total number of worldwide installed continuous dioxin monitoring systems to around 450 to 500 systems.
The increasing interest on the continuous dioxin monitoring is also supported by the work of the European TC264 WG1, which started to establish a standard as EN 1948-5 for continuous sampling of PCDD/PCDF and dioxin-like PCB.
In more than 160 applications the continuous monitoring of the dioxin emission was realized with the AMESA system.
This paper will give an overview of the experiences gained in Belgium in more than 10 years continuous sampling of PCDD/PCDF emissions mainly with the AMESA system and will present results which support the standardization work to establish EN 1948-5.

Materials and methods

The operating principles and functionality of the AMESA system were described in several publications[4,5] and have been proven through 15 years of long-term sampling of PCDD/F. In principle the used method complies with the cooled probe method of EN-1948-1 with the exception that the condensate flask is installed after the XAD-II cartridge and that therefore the condensate does not need to be collected and analysed. This is in accordance to US EPA method 23A. Additionally the plane filter for the dust collection is replaced by quartz wool included in the top of the XAD-II cartridge. The cartridge containing the adsorbed dioxins and furans is evaluated together with a data medium in an accredited laboratory. By means of this process, dioxins and furans are separated from the gas phase and the condensate in one adsorption step. With this method it is possible to collect the dioxin and furans up to 4 weeks on one XAD-II cartridge. Therefore the complete yearly dioxin emission of a plant can be determined.

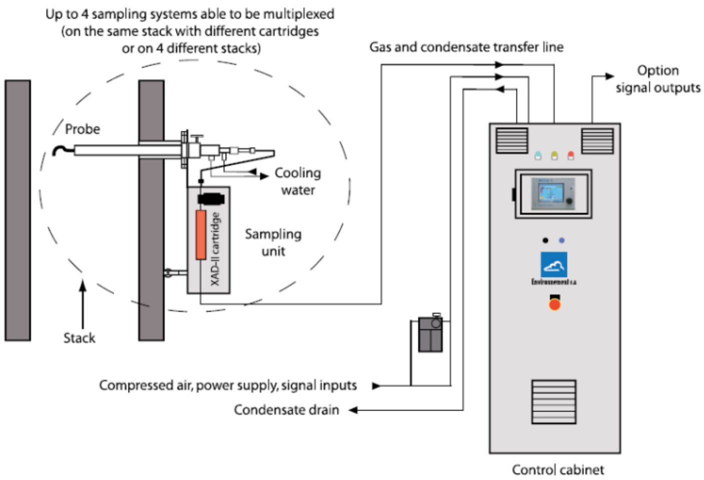

Figure 1: Functional principle of the AMESA system

In Belgium the continuous emission monitoring of PCDDs/PCDFs is regulated in Flanders Vlarem II and in Wallonia according a decree dated on 11th May 2000. According to these regulations the PCDDs/PCDFs have to be sampled in a continuous way, with at least two-weekly analyses**.** For the thus obtained results a guide value of 0,1 ng TEQ/m³ is applicable. In the meantime the requirement of 2 weeks sampling was opened to a 4 weeks sampling if the concentration are permanently low and if the operator applies for it.
From the beginning it was important to generate reliable results and therefore it was developed in Flanders and published by Vito the "Code of good practice for approval of long term dioxin sampling equipment in stacks"[6].
This document describes several requirements on the used sampling system but also the conditions when a sampling should be realized. Specific during start-up and shut-down periods of the incinerator the sampling should be interrupted, because these phases of operation do not represent standard operating conditions.
Possible criteria to decide that the plant is in operation were defined as follows:
- a maximum oxygen concentration (18 %) in the flue gases of the combustion plant
- a minimum flue gas velocity through the chimney
- a minimum temperature in the (post) combustion chamber

Other but less indicated criteria, which cannot be applied as a sole trigger for operation of the continuous dioxin sampling, were defined as:
- feeding of waste
- movement of the grates
- decision of an operator to manually switch off the sampling.

In which distribution the different criteria are used by the operators is shown in fig. 4.
In Wallonia similar requirements were defined.
The collection of data´s and the therefore generated experiences is an ongoing process. Due to this fact Vito generated an application report[7] in the year 2009 on which 13 plant operators answered and which leaded than to an update of the code of good practice, which is still in the approval phase.
As described before in the AMESA system is used a cooled probe which cools down the gas to a temperature below 50 °C. After the cooled probe the gas, condensate and dust enters directly the adsorption cartridge. For this cartridge it is not possible to approve directly the demanded dust filter efficiency of EN 1948-1 (99,5 % for an test aerosol of 0,3 um).
In several tests a back-up dust filter which fulfills the requirements of EN 1948-1 was installed downstream the adsorption cartridge (Fig. 2).
The filters were analyzed after several weeks (mainly 2 weeks) of sampling.

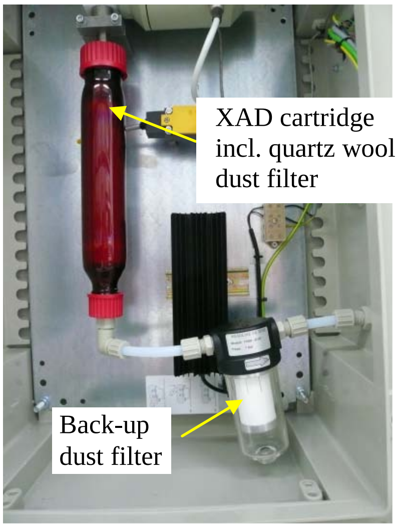

Fig. 2 XAD-cartridge with back-up dust filter

Results and discussion:
With the help of the ongoing publication of the measurement data of the Wallonia region[8] it could be demonstrated earlier, that the total dioxin emissions were reduced up to a factor of 20 in comparison to the first year of continuous dioxin monitoring, still if the quantity of burned waste increased by around 50 %.
In Flanders all the results are not in the same way available for the public like in Wallonia. However also in this region it could be demonstrated the positive effect of continuous monitoring.
Due to the use of such instruments it was also the first time possible to demonstrate the positive impact of a new flue-gas cleaning on a long term basis. E.g. in the incineration plant IVOO the dioxin emission values were reduced by a factor 5 – 10 after the installation of a catalytic DeNOx system in October 2004[9] (Fig. 3).
The results of the survey which was published by Vito in 2009 were also quite interesting. Actual all 13 plant operators who answered the questionnaire have installed in total 21 AMESA systems. On the question if the operators think that the use of a continuous dioxin sampling system is usefull 76 % answered with yes (Fig. 5).
This reflects very good the positive impression of the operators after using such systems for several years. Additional it confirms that the sceptic opinion which some operators have before the installation are not confirmed by the operation of the systems for several years. Contrary to the former doubts, the operators see as advantages the possibility to have an adequate check for the efficiency of flue gas cleaning, to have an early

warning system for disturbances, to have an alarm function with prevention of calamities and to have a proof of innocence after high dioxin depositions measured by the environment agency.

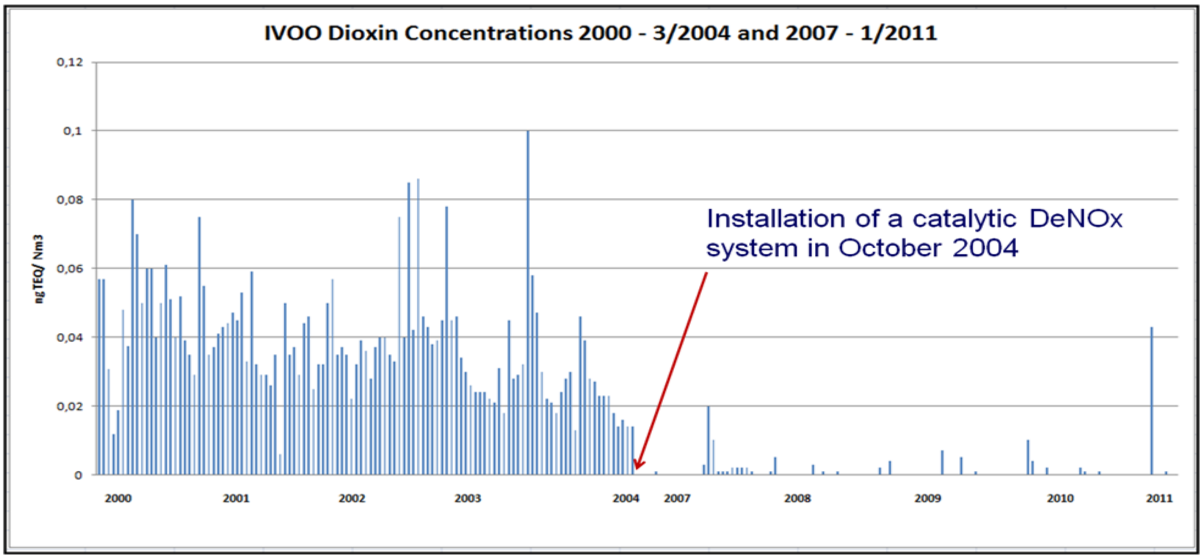

Fig. 3 Long-term (2-weeks) dioxin concentrations of the plant IVOO

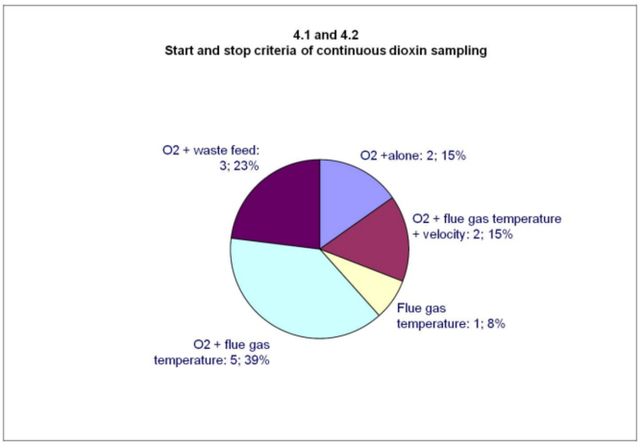

Fig. 4 Start and stop criteria in Flanders

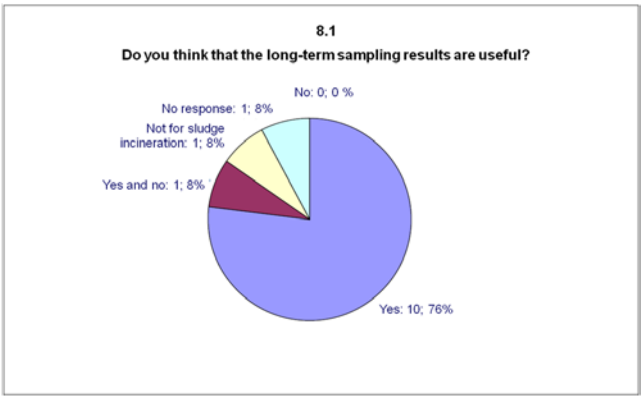

Fig. 5 Question 8.1 of the survey in Flanders

Another topic of discussion is the operating cost of such systems. In Flanders some operators complained about the high analysis costs. However as it was published for Wallonia[10] the total costs for the network of 11 stacks were in the range of 1,00 € per ton waste. These are very reasonable costs for a high benefit for the environment.

In table 1 are shown the results of the back-up filter tests. In the tested stacks the results of the dioxin analysis of the dust filters which were installed after the cartridge were for all the congeners below the limit value. Such results improve that in these stacks the dust filtering by quartz wool is sufficient and that no breakthrough of fine dust appears. This is in compliance with the experiences which were published by other groups before[11].

Filter	E4	E1	E5	E2	E6	E3
Begin date	01.09.2010	01.09.2010	15.09.2010	15.09.2010	29.09.2010	29.09.2010
End date	15.09.2010	15.09.2010	29.09.2010	29.09.2010	13.10.2010	13.10.2010
Sampled volume (Nm^3dry)	70,850	68,331	161,150	148,968	122,973	121,780
Average O2 (Vol %)	10,2	13,0	10,3	11,8	9,9	10,8
Dioxin concentrations (ng TEQ/Nm^3, dry at 11 % O2)						
XAD-cartridge	0,057	0,045	0,033	0,021	0,019	0,010
Dust filter	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

Table 1. Results of the back-up filter analysis

The recovery rates of the sampling spike standards in such tests were higher than 90 %.
These results will be presented to the WG 1 and can be used for the ongoing standardization process of EN1948-5.

References:

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