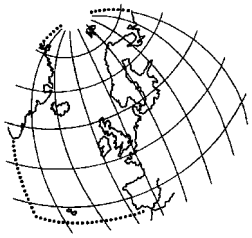
Best Available Techniques (BAT)
and Best Environmental Practice (BEP) Series

Best Environmental Practice for the Reduction or Prevention of Emissions of Polycyclic Aromatic Hydrocarbons (PAHs) from Domestic Combustion Appliances



OSPAR Commission
2001

The Convention for the Protection of the Marine Environment of the North-East Atlantic (the "OSPAR Convention") was opened for signature at the Ministerial Meeting of the former Oslo and Paris Commissions in Paris on 22 September 1992. The Convention entered into force on 25 March 1998. It has been ratified by Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxembourg, Netherlands, Norway, Portugal, Sweden, Switzerland and the United Kingdom and approved by the European Community and Spain.

La Convention pour la protection du milieu marin de l'Atlantique du Nord-Est, dite Convention OSPAR, a été ouverte à la signature à la réunion ministérielle des anciennes Commissions d'Oslo et de Paris, à Paris le 22 septembre 1992. La Convention est entrée en vigueur le 25 mars 1998. La Convention a été ratifiée par l'Allemagne, la Belgique, le Danemark, la Finlande, la France, l'Irlande, l'Islande, le Luxembourg, la Norvège, les Pays-Bas, le Portugal, le Royaume-Uni de Grande Bretagne et d'Irlande du Nord, la Suède et la Suisse et approuvée par la Communauté européenne et l'Espagne.

ISBN 0 946956 72 3

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Executive Summary

The emission of air pollutants from combustion appliances depends on combustion conditions and fuel composition. The most hazardous emissions to air from wood, peat and coal combustion are CO, CO_2, particulates (soot, tar), SO_2 and PAHs. PAHs consist of a large amount of polycyclic aromatic hydrocarbons with three or more fused rings in their molecular structure. Several different PAHs are formed in combustion processes but approximately 20 can be analysed. Usually 6-15 PAH compounds are identified and quantified using gas chromatography.

The following measures to prevent emission of PAHs from domestic solid fuel combustion appliances, are possible: the size (nominal effect) of the combustion appliances must fit the prevailing energy out-put need, although mineral fuel fired small residential heating appliances maintain their efficiency over a wide range of outputs; old combustion appliances replaced by new ones of high combustion efficiency; installation of accumulator tanks at old boilers in order to improve combustion efficiency; use of clean and dry fuel (no use of treated wood or domestic waste); use of fuel of homogenous quality and size; short start-up lighting period; frequent fuel loading in accordance with full effect operation; avoid under-loading and over-loading operations; air inlet flow adjusted to optimal combustion conditions; the chimney regularly swept and regularly maintenance/cleaning of the appliance; and soot and fly ash safely deposited.

Carbon monoxide (CO) is used as an indicator of combustion efficiency and serves as an operation parameter for combustion processes. National regulations in Denmark, Germany, Norway and Sweden all have different provisions indicating either the prohibition of using certain materials for combustion, or emissions of CO, particulates or organic gaseous carbon related to different units of e.g. fuel used or flue gas. Work is underway in CEN to establish harmonised European Standards for CO emissions on the basis of the current CEN emission guidelines for residential solid fuel stoves, cookers, boilers and open fireplaces and inserts. The Protocol on Persistent Organic Pollutants of the 1979 UNECE Convention on Long-range Transboundary Air Pollution contains requirements for the reduction of emissions of PAHs which includes emissions of PAHs from residential combustion.

Reduction of emissions of PAHs from domestic coal, peat and wood combustion appliances can be achieved in principle in two ways: installation of high quality appliances (regarding combustion efficiency) to replace the old polluting appliances, and enhancing the awareness of the users/operators regarding operation/stoking of the appliances. Emission reduction of PAHs will of course also be achieved by replacing wood or coal burning appliances by oil-/gas fired appliances or by alternative energy production systems as solar collectors.

The following measures may be relevant for the reduction of emission of PAHs: by the introduction of smoke control zones in which only smokeless fuels and smoke free appliances complying with certain standards can be used; other types of administrative actions, e.g. users guidelines, subsidies on certified stoves, negotiated agreements with manufacturers or importers, etc; and public information campaigns.

The effectiveness of the implementation of national emission reduction measures for PAHs can be monitored (or calculated) by the reduction rate of the annual emissions of PAHs from domestic solid fuel combustion appliances, as a result of replacing old combustion appliances with those certified by CEN.

Récapitulatif

Les émissions de polluants atmosphériques des appareils de combustion dépendent des conditions de la combustion et de la composition du combustible. Les émissions atmosphériques les plus dangereuses, telles que provenant de la combustion du bois, de la tourbe et du charbon, sont le CO, le CO2, les particules (suie, goudron), le SO2 et les HPA. Les HPA sont constitués par un grand nombre d'hydrocarbures polycycliques aromatiques dont la structure moléculaire comporte trois anneaux combinés ou plus. Plusieurs types de HPA se forment au cours de la combustion, et environ 20 d'entre eux peuvent être analysés. En général, l'on identifie entre 6 et 15 composés de HPA, que l'on quantifie par chromatographie en phase gazeuse.

Pour empêcher les émissions de HPA dues au brûlage des combustibles solides dans les appareils ménagers les mesures suivantes sont possibles : la taille (rendement nominal) des appareils de combustion doit impérativement correspondre à l'essentiel des besoins de production d'énergie, quoique les petits appareils de chauffage ménagers brûlant du combustible minéral conservent leur efficacité sur un large éventail de rendements ; remplacement des vieux appareils de combustion par des appareils neufs à la combustion plus efficace ; installation de réservoirs accumulateurs aux vieilles chaudières de manière à augmenter le rendement de la combustion ; utilisation de combustible propre et sec (pas de brûlage de bois traité ou d'ordures ménagères) ; utilisation d'un combustible d'une qualité et d'une taille homogène ; allumage rapide de l'appareil ; chargement fréquent en combustible pour maintenir le rendement à son maximum ; éviter de charger insuffisamment et de trop charger ; aspiration de l'air réglé pour obtenir des conditions optimales à la combustion ; ramonage régulier des cheminées et entretien/nettoyage régulier de l'appareil ; dépôt en lieu sûr de la suie et de la cendre volante.

Le monoxyde de carbone (CO) est un indicateur de l'efficacité de la combustion et est aussi un paramètre des processus de combustion. Les réglementations nationales du Danemark, de l'Allemagne, de la Norvège et de la Suède comportent des dispositions différentes, interdisant le brûlage de certains matériaux, ou indiquant des plafonds d'émission de CO, de particules ou de carbone organique à l'état gazeux en fonction de diverses unités, par exemple de combustible consommé ou de gaz perdus. Des travaux sont en cours à la CEN pour établir des normes européennes harmonisées applicables aux émissions de CO, ceci sur la base des lignes directrices CEN actuelles portant sur les émissions des appareils ménagers brûlant du combustible fossile, des cuisinières, des chaudières, des âtres et des inserts. Le Protocole sur les polluants organiques persistants de la Convention ECE de 1979 sur la pollution atmosphérique transfrontière à longue distance contient des exigences applicables à la réduction des émissions de HPA, dont les émissions de HPA des appareils ménagers de combustion.

La réduction des émissions de HPA des appareils ménagers brûlant du charbon, de la tourbe et du bois peut en principe être obtenue de deux manières : l'installation d'appareils de haute qualité (sur le plan du rendement de la combustion) afin de remplacer les vieux appareils polluants, et la sensibilisation des utilisateurs sur le plan du fonctionnement/de l'entretien de la combustion des appareils. La réduction des émissions de HPA peut bien entendu aussi être obtenue en remplaçant les appareils brûlant du bois ou du charbon par des appareils fonctionnant au mazout ou au gaz, ou par d'autres dispositifs de production d'énergie, tels que des capteurs solaires.

Les mesures suivantes sont susceptibles d'être utiles à la réduction des émissions de HPA : création de zones de contrôle des fumées, dans lesquelles seuls des combustibles et des appareils n'émettant pas de fumée, conformes à certaines normes, peuvent être utilisés ; autres types de mesures administratives, p.ex. directives aux utilisateurs, subventions aux appareils de chauffage certifiés, accord négocié avec les fabricants ou les importateurs, etc ; et campagnes d'information de la population.

L'efficacité de la mise en œuvre des mesures nationales de réduction des émissions de HPA peut être vérifiée (ou calculée) par le taux de réduction des émissions annuelles de HPA des appareils ménagers brûlant des combustibles solides, ceci en conséquence du remplacement des vieux appareils par des appareils certifiés par le CEN.

1. Emission reduction potential of domestic coal, peat and wood combustion

1.1 Introduction

1. The emission of air pollutants from combustion processes depends on the combustion conditions and the fuel composition. The emission of PAHs is strongly correlated to the combustion quality of the appliance technology used, and the operation conditions including the fuel quality. Normally the amount of PAHs emitted per unit of energy output from small combustion appliances is several orders of magnitude higher than from larger appliances. Good combustion conditions are easier to achieve in large combustors and abatement systems are also then often in place.

2. The most hazardous emissions to air from pure wood combustion are CO, particulate (soot, tar) and PAHs. The most hazardous emissions from coal combustion are CO, CO_2, particulates, PAHs and SO_2.

3. Comparison of PAH emission data from small stoves and fireplaces must be carried out very carefully because such data include a relatively high uncertainty due to emission variations during the combustion cycle and the complex sampling and analytical methods.

1.2 PAHs

4. PAHs consist of a large number of polycyclic aromatic hydrocarbons, i.e. hydrocarbons with 3 or more fused aromatic rings in their molecular structure. Several different PAH compounds are formed in combustion processes, but approximately 20 can be chemically analysed. Normally 6 - 15 PAH compounds are identified and quantified using chromatographic analytical methods.

1.3 Combustion appliances and the combustion process

5. Domestic combustion appliances, boilers, usually consist of a combustion chamber, primary and secondary air supply and a heat exchange system. The heat exchange systems in residential boilers tend to be water based. Small stoves and open fireplaces do not have a water based heat exchange system, but the heat is directly radiated into the indoor air. Domestic combustion appliances do not usually have filters or other abatement systems installed.

6. In the combustion chamber the fuel undergoes drying, pyrolysis / degassing and finally the combustion process when volatile carbon containing compounds in the flue gas react with air oxygen (oxidising) to form the end products CO_2 and H_2O. Most impurities (e.g. sulphur) in the fuel undergo similar oxidising processes forming oxidised end products, e.g. SO_2. These reactions occur mainly in the primary combustion chamber. However, the oxidation processes are never complete, and therefore bigger appliances are equipped with a secondary combustion chamber where the remaining unreacted compounds can undergo a final oxidation step, either a thermal or a catalytic oxidation.

7. The PAHs are formed in the combustion process from unreacted carbon compounds in the hot flue gas at temperatures of about 500 - 800 °C. When the flue gas cools down, the PAH components condense from the vapour phase onto particles (1). The heavier and most carcinogenic PAH components (PAH components containing 4 benzene rings or more) are normally emitted while adsorbed on soot and dust particles. Because of the relatively larger surface of small particles, the PAH concentrations on small particles are higher than on large particles. Flue gas temperatures in small domestic appliances rarely exceed 500^oC. Temperatures in the fuel bed may be up to 1500^oC and overbed temperatures in the range 500 to 800^oC at full output. At low output these temperatures are very much lower and probably all below the 500 to 800^oC required for the formation of PAHs.

8. The formation and emission of PAHs are highly dependent on the combustion conditions, and consequently also on the design of the combustion appliance and the technology used, and on the loading operation. Efficient combustion can only be achieved when the temperatures in the combustion chamber are high enough, and when flue gases have good access to air oxygen and enough time to undergo the combustion (oxidation) reactions. The hot flue gases do not easily mix with the colder inlet air. Therefore the combustion appliances must be designed with special focus on achieving optimal flue gas-air mixing conditions and the combustion chamber must allow a long enough flue gas residential time.

9. There are in principle three types of small wood stoves on the market;

- one-chamber stoves with or without air preheating;
- combustion appliances equipped with a secondary combustion chamber;
- combustion appliances equipped with catalysts.

10. Domestic boilers can be categorised according to the stoking technology used, into the following groups:

- hand stoked room heaters;
- hand stoked heating boilers;
- automatically stoked heating boilers.

11. The size (nominal effect, energy output) of a combustion appliance should be chosen in accordance with the energy output needed, because overloading or underloading of stoves or boilers affects the combustion conditions negatively, thus increasing the emissions of PAHs.

1.4 Fuel

12. The combustion quality is also dependent on the type and quality of the fuel. In general it is more difficult to achieve good combustion conditions when using solid fuel compared to liquid or gaseous fuel.

13. To achieve good combustion conditions when using solid fuel, the fuel must be dry to avoid energy losses due to water evaporation. To some extent moisture will promote equal combustion conditions. Its negative effect on combustion efficiency is solely due to the energy required for evaporation and this amounts to about 1% reduction for each 10% of moisture content. Wood containing a too high moisture content can cause of deposits in chimneys. Such deposits indicate the use of unconditioned or “green” fuel.

14. In order to achieve a smooth loading as possible (see Operation conditions), the size of the fuel units (wood logs, briquettes, etc) should be adjusted in accordance with the combustion appliance requirements. In general frequent loads of small sized fuel units disturb the combustion process less than infrequent loads with large sized fuel units.

Table 1: Typical content of wood, peat and coal fuel is (2, 8):

	Wood*	**Peat***	**Coal**
Element / compound content			
Hydrogen (%)	6	5,0 – 6,0	5,5
Carbon (%)	50	50 - 60	83
Oxygen (%)	42	33	9
Ash (%)	1 - 3	5,3	5 – 20
Sulphur (%)	0,04	0,1 – 0,3	1
Nitrogen (%)	0,2	0,5 – 3,0	1,5
Chlorine (%)	0,0045	0,02 – 0,06	0,2
Mercury (ppm***)	0,02	0,08	0,3
Cadmium (ppm***)	0,18	0,13	1
Lead (ppm***)	3,6	5,1	15
Water (%)	20	50	3 – 10
*Energy content (MJ**/kg)*	14 - 15	10 -14	28

* dry fuel ** MJ = Mega Joule = 10^6 Joule *** ppm = mg/kg

1.5 Emissions of PAHs

15. PAHs as well as CO are formed as a consequence of incomplete combustion (oxidation) processes. Therefore the concentration of PAHs in the flue gas is closely related to the CO concentration. CO is normally used as one of the main emission parameters.

16. In the combustion process all types of polycyclic aromatic hydrocarbons (PAHs) as well as bicyclic aromatic compounds (naphthalene, etc.) and other organic matter are formed. The PAH profile in flue gases from combustion processes is rather similar, regardless of the fuel and the technology used. Normally phenanthrene is the most dominating compound accounting for approximately 40% of the total emission of PAHs. The carcinogenic PAH component benzo(a)pyrene accounts for approximately <1 - 5%.

17. In the flue gas the heavier PAH compounds (PAH components containing 4 benzene rings or more) are semi-volatile gases. They are therefore partly or totally adsorbed on dust or soot particles at the emission point, depending on the flue gas temperature.

18. According to a Norwegian study the particles emitted from small wood stoves and an open fireplaces mainly consist of tar (3). The PAH content of the tar varies depending of the course of the pyrolytic processes in the combustion. According to the study mentioned above, emissions of PAHs (14 compounds) accounted for <0,1 - 0,6% of the tar emission.

19. The emissions of particles and PAHs during the start up period is normally several orders of magnitude higher than during stable operation conditions. From a tiled wood stove approximately 80% of the total amount of PAHs emitted during a combustion period lasting 2 hours, was emitted during the first 15 minutes (4). The emission of PAHs decreases when stable combustion conditions have been reached. During the burnout period the emission of PAHs is normally very small, in contrast to the CO emission.

20. Emissions of PAHs from small combustion units can be considerable unless the combustion conditions are optimised. Normally the emission of PAHs expressed as unit of weight per unit of energy input (mg /MJ_{input}) is much higher for small combustion units compared to larger ones.

21. The following PAH emissions have been measured:

	PAH-emissions (amount/energy input)	**Ref.**
Wood combustion		
residential heating boilers (11- 40 kW)	<1 - 24 mg/MJ (16 PAH)	5
residential heating boiler, stoker screw (23 kW)	0,1 - 17 mg/MJ (35 PAH)	6
wood stoves (approx. 15 kW)	0,6 - 9 mg/MJ (26 PAH)	3
Coal combustion		
residential heating boiler, stoker screw (23 kW)	approx. 0,2 mg/MJ (35 PAH)	6

22. The PAH emission factors for coal and wood combustion in small stoves and fireplaces have been proposed to be approximately 0,3 mg/kg (approximately 0,02 mg/MJ) and 40 mg/kg (approximately 3 mg/MJ) respectively (7).

23. In general emission of PAHs from domestic coal combustion during stable operation is lower than from wood combustion appliances, due to higher combustion temperatures and more homogenous fuel.

1.6 Operation conditions

24. The emission of CO, as an indicator of combustion efficiency, is used as an operation parameter for combustion processes. However, small combustion appliances are normally not equipped with CO monitoring systems. The combustion efficiency can therefore not be observed during the combustion operation. The operator must have enough general information about combustion processes in order to make operational adjustments (stoking) to optimise the combustion conditions.

25. The most important operational parameters are the size, quality and quantity of the fuel (see Fuel), the inlet airflow and the fuel loading frequency.

26. The combustion conditions are very dependent on the oxygen available for the combustion (oxidation) reactions and therefore the inlet airflow into the combustion chamber must be carefully adjusted. When operating under starved conditions the oxygen deficit prevents the combustion reactions to be completed. When operating with air excess, the combustion chamber will cool, thus decreasing the combustion efficiency. Both these operation modes lead to increased emission of PAHs, often several orders of magnitude higher than under stable and most efficient conditions.

27. Every load of solid fuel disturbs the conditions in the combustion chamber negatively, resulting in periodically increased emission of PAHs. The more "running" loading, the easier it is to achieve a stable and an efficient combustion. In principle the fuel loads should be rather small and the loading rather frequent. By automatically loading systems (stoker screw, etc) improved combustion conditions can be achieved.

1.7 Waste products

28. The waste products from wood and coal combustion consist of bottom ash (slag) and fly ash (soot, tar and carbonised particles). Bottom ash remaining from efficient combustion is normally mineralised containing very little carbon and negligible amounts of PAHs. In some countries bottom ash from biomass combustion is used as fertiliser in forests.

29. Fly ash may contain acid components, metals and organic compounds, e.g. PAHs. Fly ash and soot should therefore be safely deposited.

1.8 Maintenance

30. Bottom ash from combustion unit should be regularly removed. The chimney should be regularly swept in order to prevent accumulation of PAHs, tar and soot, that can result in re-emission of pollutants, e.g. PAHs, and can initiate chimney fires.

1.9 Emission reduction measures

31. For practical purposes the following measures to prevent emission of PAHs from domestic solid fuel combustion appliances, are possible:

- the size (nominal effect) of the combustion appliances must fit the prevailing energy out-put need, although mineral fuel fired small residential heating appliances maintain their efficiency over a wide range of outputs;
- old combustion appliances replaced by new ones of high combustion efficiency;
- installation of accumulator tanks at old boilers in order to improve combustion efficiency;
- use of clean and dry fuel (no use of treated wood or domestic waste);
- use of fuel of homogenous quality and size;
- short start-up lighting period;
- frequent fuel loading in accordance with full effect operation;
- avoid under-loading and over-loading operations;
- air inlet flow adjusted to optimal combustion conditions;
- the chimney regularly swept and regularly maintenance/cleaning of the appliance;
- soot and fly ash safely deposited.

2. Implementation of reduction measures

2.1 Existing national regulations and quality control

32. In order to reduce emissions from small combustion appliances, which represents a considerable local pollution source especially in the cold season, some countries have introduced wood stove regulations.

33. In Norway a Regulation was introduced in 1998, which requires that only certified new wood stoves can be installed in buildings. A wood stove quality test has been established. A weighted limit value of 10 g particles/kg dry fuel has been set for emissions from traditional wood stovesand 5 g particles/kg dry fuel for catalytic stoves.

34. In Sweden new installations in densely built areas are regulated as follows (binding rules): Residential biomass heated boilers (<50 kW) must not emit more than 150 mg OGC (organic gaseous carbon)/ Nm^3 dry gas at 10% O_2, and wood stoves and inserts must not emit more than 250 mg OGC/Nm^3 dry gas at 13% O_2,. There are non-binding recommendations for biomass boilers > 50 kW: dust max. 350 mg/Nm^3 at 13% CO_2; CO max. 500 mg/Nm^3 dry gas as a mean value.

35. In Denmark regulations on power production require that only natural wood be used in wood stoves. Use of any kind of waste materials e.g. MDF-board, chipboard and wood, that is painted or preserved, is banned. No general emission standard applies to wood stoves. Emissions from domestic solid fuel fired installations are regulated by provisions in the Environmental Protection Act §42 and are enforced by the Municipal Councils. Furthermore Denmark has recently finished investigations on low power-high efficiency wood stoves. The report is available on URL:
(http://www.mst.dk/Default.asp?Sub=/udgiv/publikationer/2000/87-7909-792-8html/).

36. In Germany emission standards for domestic solid fuel fired installations are laid down in the Ordinance on Small Firing Installations (1st BlmSchV). This ordinance requires that firing installations with normal heating capacity exceeding 15 kW, mostly heating boilers, shall be constructed and operated in such a way that the particulate emissions do not exceed the following limits values: 0,15 g/m^3 (normalised to 13% O_2) for burning peat, hard-coals, and lignite; 0,15 g/m^3 (normalised to 13% O_2) for burning natural wood, wood pellets and briquettes, and straw. More stringent emission limit values for CO have been set in accordance with increased heat capacities of the appliances:

Heat capacity	CO limit value g/m^3 (norm. 8% O_2)
15 – 50 kW (100 kW for straw combustion):	4
> 50 – 150 kW:	2
> 150 – 500 kW:	1
> 500 – 1000 kW:	0,5

37. In the USA (EPA) wood stove emission have been regulated by law. In Canada and Australia wood stove emission regulations have been proposed.

38. The Protocol to the 1979 UNECE Convention on Long-range Transboundary Air Pollution on Persistent Organic Pollutants, and the Executive Body Decision 1998/2 (ECE/EB.AIR/60) contains requirements on emission reduction of PAHs from main emission sources. PAH is defined as the sum of benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, and indeno(1,2,3-cd)pyrene. The protocol includes emissions of PAHs from residential combustion.

39. In order to implement the best available technique concept also for small combustion units the European Committee for Standardisation (CEN) has established emission guidelines for residential solid fuel stoves, cookers, boilers and open fire places and inserts. The four CEN European Standards (ENs), four various types of combustion appliances now approved for publication after Formal Vote, use the measurement of carbon monoxide as the bellwether of good combustion, i.e. if the carbon monoxide is low then the other constituents of the flue gases will be low and the combustion process efficient. This parameter is chosen because it is easy to measure compared to PAH and particulates. CEN/TC 295 is now working on the conversion of the four ENs to harmonised hENs under the Mandate M/129 of the CPD. First drafts have already been circulated to CEN/TC 295 (Stage 32) and are planned to reach Stage 49 (Document available for Formal Vote) by 28th February 2002.

40. The European Commission has set up a working group to review the available evidence and recommend a possible limit value for PAHs. The group is expected to report shortly. The Commission will then prepare a proposal for a further daughter directive on air quality, which could be adopted by mid/late 2002. Present signs are that the working group will recommend that toxicological data would support a limit value, possibly between a range of values to be achieved by 2010.

2.2 Choice of action

41. Reduction of emissions of PAHs from domestic coal, peat and wood combustion appliances can be achieved in principle in two ways: installation of high quality appliances (regarding combustion efficiency) to replace the old polluting appliances, and enhancing the awareness of the users/operators regarding operation/stoking of the appliances. Emission reduction of PAHs will of course also be achieved by replacing wood or coal burning appliances by oil-/gas fired appliances or by alternative energy production systems as solar collectors.

42. The following measures may be relevant for the reduction of emission of PAHs:

- by the introduction of smoke control zones in which only smokeless fuels and smoke free appliances complying with certain standards can be used;

- other types of administrative actions, e.g. users guidelines, subsidies on certified stoves, negotiated agreements with manufacturers or importers, etc;
- public information campaigns.

2.3 Monitoring of the effectiveness of the implementation of an OSPAR Recommendation

43. The effectiveness of the implementation of national emission reduction measures for PAHs can be monitored (or calculated) by the reduction rate of the annual emissions of PAHs from domestic solid fuel combustion appliances, as a result of replacing old combustion appliances with those certified by CEN.

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ISBN 0 946956 72 3