Comments filed to U.S. EPA draft report "Emissions Inventory of Section 112(c)(6) Pollutants: Polycyclic Organic Matter (POM), 2,3,7,8-Tetrachlorodibenzo-p-dioxin (TCDD)/2,3,7,8-Tetrachlorodibenzofuran (TCDF), Polychlorinated Biphenyl Compounds (PCBs), Hexachlorobenzene, Mercury, and Alkylated Lead"

Review of the Polycyclic Organic Matter Emission Inventory for Residential Wood Combustion

U.S. Environmental Protection Agency Draft Report:

Emissions Inventory of Section 112(c)(6) Pollutants: Polycyclic Organic Matter (POM), 2,3,7,8-Tetrachlorodibenzo-p-dioxin (TCDD)/2,3,7,8-Tetrachlorodibenzofuran (TCDF), Polychlorinated Biphenyl Compounds (PCBs), Hexachlorobenzene, Mercury, and Alkylated Lead

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Introduction

The Hearth Products Association (HPA) has contracted AGI Technologies as an independent consultant to review the U.S. Environmental Protection Agency's (EPA) Section (112)(c)(6) emission inventory draft report. The review focused on emissions of polycyclic organic matter (POM) attributed to residential wood combustion (RWC). Based on the review it is concluded that: 1) The parameters (7-PAH, 16-PAH and EOM) used to assess POM emissions are not acceptable surrogates for total POM, 2) The number of actual measurements on which emission factors are based are grossly inadequate to provide values representative of the entire population of RWC devices in use, 3) The quality of the relatively few actual measurements of emissions factors are low, and the method used to calculate overall RWC emission factors from them is flawed, and, 4) An erroneously large national level activity value for RWC was used to calculate total national emissions. A discussion of each of these four points follows.

7-PAH, 16-PAH and EOM as Surrogates for Total POM

The Clean Air Act Amendments (CAAA) of 1990 requires the identification of sources responsible for a least 90% of POM (and six other pollutants). In order to meet this requirement, the stated intent of EPA's draft report is to present results of a national emission inventory of POM (along with the six other pollutants). This was not done. Instead, national emission inventories for seven specific polycyclic aromatic hydrocarbons (7-PAH), sixteen specific polycyclic aromatic hydrocarbons (16-PAH), and extractable organic material (EOM) were presented in lieu of an emission inventory for total POM. None of these three parameters are good surrogates for total POM.

Polycyclic aromatic hydrocarbons (PAH) as a group is a subset of polycyclic organic material (POM), furthermore, the seven and 16 specific polycyclic aromatic hydrocarbon compounds (7-PAH and 16-PAH) comprise a very small subset of the total number of PAH. The emission factors for 7-PAH and 16-PAH listed in the EPA draft report are taken from the EPA "Locating and Estimating Air Emission" (L & E) series (reference 1), which in turn takes the values from EPA's AP-42 compilations (reference 2) and two other older reports (references 3 and 4). The EPA draft report states, "The complex mixture of POM consists of literally thousands of organic compounds." The L & E document states, "Theoretically, millions of POM could be formed."

It is widely recognized that the concentration of specific chemical compounds that make up the POM fraction of emissions from various air pollutant source categories (e.g., vehicular exhaust versus RWC) vary widely. This can be seen in reviewing the relative concentration of the PAH compounds for different source categories tabulated in the L & E document. In fact, the documented variability of specific PAH species from source category to source category has been suggested as a tool to apportion the relative contribution of different pollutant source categories to measured ambient concentrations (references 5 and 6). For example, in reference 5, it was concluded that the relative proportion of specific PAH species vary over several orders of magnitude from different source types.

The facts that: 1) Seven and 16 specific compounds are being used as indicators of total POM levels which are made up of thousands to millions of different compounds, and, 2) The relative fraction of total POM made up of specific compounds vary widely from source category to source category, demonstrate that the percentage contribution by source for 7-PAH and 16-PAH (shown in Figures 3-1 and 3-2 in the EPA draft report) are not representative of the percentage contribution by source for POM. In light of these concerns selecting a different set of 7 (or 16) PAH as "surrogates" for total POM may have resulted in a different list of significant source categories. It certainly could have resulted in different estimates of the magnitude of total POM emissions from candidate source categories, and, therefore, a different ranking of the relative importance of particular source categories.

The use of EOM as a surrogate for POM is inappropriate. Much of the organic material collected by emission sampling equipment will contribute to the EOM value. Emissions from a source could have a high EOM value without any POM present at all. EOM is simply a measure (an inexact measure) of organic compounds with a low vapor pressure and that can be put into solution with solvent. For this reason, RWC shows a high EOM value as compared to many other source categories. Numerous measurements of the organic carbon, elemental carbon and inorganic content of RWC emissions have shown that more than 80% of particulate emissions

(which includes the condensable fraction) are made up of organic compounds (references 6-8). For RWC the overwhelming majority of these compounds are oxygenated aliphatic and monoaromatic compounds not POM. (references 9-11). As with 7-PAH and 16-PAH, the relative percentages shown for EOM by source category in the EPA draft report (Figure 3-3) are not representative of the relative percentages of POM, moreover. in the case of RWC, the EOM value is high simply because RWC emissions are very high in non-POM organic compounds that will show up as EOM.

RWC Data Base Size

There were 22.9 million households in the United States which used wood for heat in 1990 (reference 12). Some of these households have more than one wood burning device. The 22.9 million household value is based on a survey that assigned it a 10.1% relative standard error (RSE). Based on the facts that some homes have more than one wood burning device (e.g., both a fireplace and a woodstove) and that the survey value has a 10.1% RSE, a reasonable estimate of the total number of wood burning devices in the United States in 1990 would be 25 million. A supplement report to the Household Energy Consumption Survey (reference 13) reported 8.4 million households burned more than one cord of wood per year and 14.5 million households burned less than one cord of wood per year. Most frequently homes which burn more than one cord of wood would primarily be using a woodstove and those burning less than one cord of wood would be using a fireplace. If wood furnaces are grouped with woodstoves and one takes into account that about 7% of the households that use wood as a primary source of heat use a fireplace (references 13 and 14), and that some homes may have more than one wood burning device, a reasonable estimate of the total number of woodstoves and fireplaces in the United States in 1990 would be nine million and 16 million, respectively. Based on wood use data (references 12-14) it can be estimated that about 21% of the total cordwood burned in the United States was burned in fireplaces and 79% in woodstoves. These estimates compare favorably with the estimates provided in Appendix A of the EPA draft report (28% for fireplaces and 72% for woodstoves). It also should be noted, as will be discussed later, that the HPA believes that no more than about 5% of the woodstoves in use in 1990 were new technology catalytic/non-catalytic stoves.

The EOM emission factor for all wood burning devices listed in the EPA draft report is based on only 14 tests (reference 11). In Appendix A of the EPA draft report it erroneously states that the EOM emission factor is based on tests with 12 conventional woodstoves and two catalytic woodstoves. This is not correct. One conventional woodstove (a Scott brand stove) was used and tested under 12 operating conditions and one catalytic woodstove (an Earth brand stove) was used and tested under two operating conditions (see reference 15 for test conditions). This error is understandable as reference 11 did not cite the primary report (reference 15) which described the tests. The 7-PAH and 16-PAH emission factors for conventional woodstoves listed in the EPA draft report are taken by AP-42 from the same study (reference 15), while the emission factor for high technology catalytic/non-catalytic woodstoves were based on six additional studies referenced in AP-42. According to the L & E document, "There are fewer PAH emissions test data for fireplaces as compared to woodstoves." Of the two references cited in the L & E document for fireplace tests, the tests listed in one of the references (reference 4) are described in detail in reference 9. The tests are comprised of sampling a single fireplace two times for PAH (two wood types). The second fireplace reference cited in the L & E document (reference 3) is a 1980 literature review of work conducted in the 1970's. The method used to develop the emission factors are not documented in the review but they appear to be a non-statistical "blend" of fireplace test results from three studies including the one fireplace study reported in references 4 and 9.

In summary, the data base for emission factors is not adequate. The EOM emission factor for all 25 million wood burning devices is based on one conventional and one catalytic stove. The 7-PAH and 16-PAH emission factors for the eight to nine million conventional woodstoves, which are responsible for the overwhelming majority of wood consumption and POM emissions, are based on one woodstove. The 7-PAH and 16-PAH emission factors for the 16 million fireplaces are based on no more than several fireplaces tested in the 1970's. (Documentation on tests on only one fireplace have been definitively identified.) There appear to be a few more tests available for high technology catalytic/non-catalytic woodstoves; however, since they represented a

relatively small fraction of the total woodstoves in use in 1990 and their emission factors are smaller than conventional stoves, a detailed accounting of the origins of the 7-PAH and 16-PAH emission factors was not conducted in this review.

Basing emission factors on a limited number of tests is a more serious problem for RWC than most other sources of POM because of the very high variability that can be expected for POM emissions from RWC. It has been well documented that combustion conditions such as temperature, available oxygen, and residence time will influence the production of POM (reference 16). There are many hundreds of types or models of wood burning devices in use, many dozens of tree species are commonly used for wood fuel, draft characteristics vary from home to home (chimney conditions), household altitude is variable, there are variations in fuel wood seasoning and storage practices (wood moisture), and there are wide variations in home owner operation of a wood burning devices (burn rate, burn duration, damper setting, kindling approach, etc.). Each of these parameters have significant impacts on combustion conditions and will impact POM emissions. Beyond the variability in woodstove emissions which is due primarily to the differences in combustion conditions and has been well documented for other air pollutants such as particles and carbon monoxide, the variability in the chemical makeup of wood is an additional source of variability for POM as specific POM compounds will be formed by the rearrangement and combining of compounds contained in the wood fuel. Wood is composed of lignin, cellulose, hemicelluloses, and resins. The ratios of these major chemical groups vary from tree species to tree species, particularly in wood from deciduous versus coniferous trees. Resin content, for example, may be particularly important as resins are composed of polyaromatic structures.

To provide insight into the variability of POM emissions associated with RWC, the reader is referred to reference 15 which provides the basis for the EOM emission factor for all 25 million wood burning appliances and the 7-PAH and 16-PAH emission factors for somewhere between eight and nine million conventional woodstoves. Emissions from a single conventional woodstove (a Scott brand stove) and a single catalytic woodstove (an Earth brand stove) were measured. There were twelve tests performed on the conventional woodstove. Two replicate runs were performed each on low and high burn rates using pine fuel at both high and low altitude, and two replicate runs were performed on low and high burn rates using oak fuel at low altitude (six sets of conditions with two replicate runs each). The mean EOM, 7-PAH and 16-PAH emission factors for conventional stoves from these tests are 23.4 lbs/ton, 0.051 lbs/ton and 0.69 lbs/ton, respectively. (The values tabulated in the EPA draft report are a little lower because the data from the two catalytic stove tests were included in the mean values presented there.) The standard deviation around the EOM, 7-PAH and 16-PAH means are 19.5 lbs/ton, 0.052 lbs/ton and 0.42 lbs/ton, respectively. These standard deviations represent 83%, 102% and 61% of the means for the EOM, 7-PAH and 16-PAH values, respectively. It must be emphasized that these values are for a single stove tested 12 times with two fuels, two altitudes, and two burn rates and that one half of the test were replicate tests. The magnitude of the variation in POM emissions among the very large number of parameters encountered among the real-world use of RWC devices must be very large. Statistically using one conventional stove and several fireplaces to represent millions of devices is fundamentally in error.

Quality of Emission Factors

All emission factors listed in the L & E document which is the source of the 7-PAH and 16-PAH emission factors used in the EPA draft report have an emission factor rating of E except for the one fireplace data set which was derived from a 1980 literature review article. The emission factor rating for that data set is U5. An E rating is the lowest and is described as "poor", the U rating is defined as unrated or unratable. The U5 subcategory is further defined as having a "lack of supporting documentation."

The EOM emission factor developed from the 12 tests on a single conventional stove and on two tests on a catalytic stove were obtained by using a non-reference, non-standard sampling protocol. (One author of this

review was also the co-author of the study on which the EOM emission factor is based C reference 15.) An aliquot of solvent extracts from filters, extracts from XAD-2 resin and probe rinses underwent gravimetric and chromatographic analyses. The EOM value is the sum of the gravimetric and chromatographic determinations on each of the three solutions. It is the authors opinion that the propagated uncertainty of the technique and subsequent addition of six values produced a precision of no better than 30%. Also the gravimetric sample was lost for one run reducing the EOM data set to 13.

Beyond the accuracy and precision of the fundamental measurements, the HPA is concerned about how the emission factors for the various wood burning devices were weighted to produce overall emission factors that were subsequently multiplied by the total national wood use to obtain total RWC values for the national emission inventory tabulation. For example, the L & E document from which the EPA draft report took the weighted 7-PAH and 16-PAH emission factors has tabulations for conventional woodstoves (Table 4.1-1), non-catalytic woodstoves (Table 4.1-2), catalytic woodstoves (Table 4.1-3), pellet stoves (Table 4.1-4), and fireplaces (Table 4.1-5). The emission factors of 0.035 lbs/ton for 7-PAH and 0.517 lbs/ton for 16-PAH for residential wood combustion are listed in appendices A and B of the L & E document and in appendix B and in Table 3-1 of the EPA draft report without any explanation of the calculations used to derive them. Apparently, a weighing factor was used to account for the relative usage of the various devices. In addition, the PAH data for fireplaces shown in Table 4.1-5 of the L & E document are missing a number of the 7-PAH and 16-PAH compounds. There is no explanation on how this lack of data was treated in calculating the weighted emission factors. The Household Energy Consumption and Expenditures survey results (reference 12) also show that 3.5 million cords were burned in other wood burning devices (primarily wood furnaces). No weighing or emission factors have been presented for them.

The development of the EOM emission factor for residential wood combustion is addressed in section A.24 of appendix A of the EPA draft report. In that section it is stated,

> *"Statistical data from a 1990 annual survey of residential homeowner use conducted by the EIA were used to develop the weighing factors to apply to the available emission factor data to represent the split between woodstove and fireplace use. The average nationwide percentage of wood consumption is 28 percent for fireplaces and 72 percent for woodstoves. Consumption for woodstoves can be further divided into approximately 70 percent conventional woodstoves (no control devices) and 30 percent catalytic/non-catalytic woodstoves."*

There are two key issues that need to be addressed in regards to these statements. First, the data in the referenced EIA report do not provide a direct mechanism to calculate the relative wood usage between fireplaces and woodstoves. As discussed earlier, the 28% to 72% split for wood use between fireplaces appears reasonable. However, it is not a rigorous quantitative number that can be used to calculate weighted emission factors. (It is unclear whether this split was used to calculate weighted 7-PAH and 16-PAH emission factors as discussed in the preceding paragraph.) The second issue is that the estimate of 30% wood use in catalytic/non-catalytic woodstoves in 1990 is too high. Under federal regulations conventional woodstoves could be manufactured up to July 1, 1988 and sold up to July 1, 1990. While many manufacturers started manufacturing and selling Phase I (and Phase II) certified catalytic and non-catalytic woodstoves prior to these cutoff dates, 30% wood use in them during 1990 is not a reasonable estimate based on the turnover rate of woodstoves. It is likely that less than 5% of the woodstoves in use in 1990 would have been catalytic/non-catalytic stoves. Wood use attributed to them as compared to conventional woodstoves, would be more or less at the same percentage, since they are on one hand, more efficient but on the other hand, one might argue more serious wood burners would purchase them. The low estimate is confirmed by the results of a survey conducted by the Oregon Department of Environmental Quality in Portland, Oregon for 1993 (reference 17). The results showed that 35% of the homes with woodstoves and stove-like inserts had certified devices in 1993. Certified devices in this case included both Oregon and EPA certified devices. The state of Oregon did not allow conventional stoves to be sold retail after July 1, 1986. (Oregon certification was, by in large, the model on which EPA certification was subsequently based). Consequently, in Portland, Oregon, homeowners took six and one-half years to replace 35% of their woodstoves with certified stoves. Again, nationwide (except for

Oregon) conventional woodstoves could be sold up to July 1, 1990 and, of course, the base year for the EPA draft report is 1990.

It is further stated in section A.24 of Appendix A that,

> *"Table A-16 lists the emission factors for each pollutant that were used in the inventory. The EOM factor is a weighted emission factor which represents conventional and catalytic/non-catalytic woodstove use. The EOM emissions factor was developed from test results for 14 woodstoves; 12 of these were conventional stoves and the other 2 were catalytic designs. The EOM emission factor represents an average of these test results weighted based on the percentage of conventional and catalytic/non-catalytic woodstove use described above."*

There are three points that need to be addressed regarding these statements. First, as previously discussed, the EOM emission factor was developed from 12 tests on a single conventional woodstove and on two tests with a single catalytic woodstove which will effect the weighing calculation. Secondly, also as previously discussed, a 30% catalytic/non-catalytic number is too high for 1990, and, third, the EOM emission factor (18.66 lbs/ton) listed in Table A-16 of Appendix A was multiplied by 45.6 million tons (rounded off and reported as 46 million tons in Appendix A) to produce a total emission value for RWC of 425,448 tons/yr (Table 3-2 and Figure 3-3 of the EPA draft report). No weighing was performed for the 28% of wood purported by the EPA draft report to be used by fireplaces.

National Level Activity Value

The total cordwood usage value of 45.6 million dry tons for 1994 was reported in appendix B of the L & E document and in appendices A and B of the EPA draft report (rounded off and reported as 46 million in appendix A). This value was multiplied by the 7-PAH, 16-PAH and EOM emission factors to obtain the total RWC values for 1990 of 800 tons/yr, 11,800 tons/yr and 425,448 tons/yr, respectively.

According to the Household Energy Consumption and Expenditure survey (references 12 and 13), 29.1 million cords of wood were burned in 1990 (actually December 1989 through November 1990). The Energy Information Administration uses a conversion factor of 1.163 tons per dry cord (reference 14) which is consistent with the mean cord weight of 1.212 tons per dry cord determined for 36 tree species (standard deviation around the mean of 0.386 tons) commonly used as fuel (reference 18). The 29.1 million cords of wood burned in 1990 multiplied by the conversion factor of 1.163 tons per dry cord yields 33.8 million tons of wood burned in 1990, a significantly lower value than the value (45.6 million tons) used by the EPA. Additional confusion regarding the correct national level activity value appears to be associated with the facts that both appendices A and B of the EPA draft report and appendix B of the L & E document cite a document that does not contain the national activity level (reference 19 here), and the Energy Information Administration states that the wood energy consumption originally reported for 1990 as 786 trillion BTU was subsequently revised to 581 trillion BTU (see Table 1, page 16 of reference 14). The number of tons of wood corresponding to 786 and 581 trillion BTU are 45.6 and 33.8 million, respectively.

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