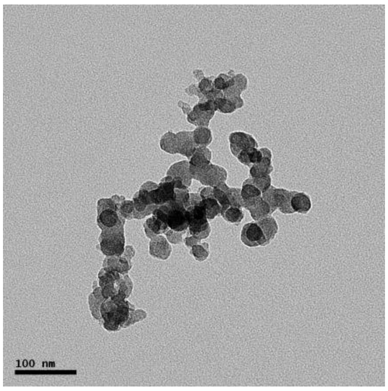
Emission inventory of black carbon for the Helsinki Metropolitan Area in 2010



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Research report

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Measurement, Monitoring and Environmental Assessment (MMEA)

WP4.5.2 Advanced methods to study fine particle chemistry in combustion

Outline

1. Introduction

Black carbon is a product of incomplete combustion and is emitted as solid particles. Black carbon is one of the few particulate specie that absorbs solar radiation. The primary BC particles are small spherules of around 10 nanometers in diameter that form agglomerates 100 to 1000 nm in diameter. Carbonaceous aerosols are constituents of airborne particulate matter, ao. $PM_{2.5}$, and are also a concern due to their relatively high ambient concentrations and subsequent health effects. Since black carbon is almost solely composed of carbon atoms, the term elemental carbon is often used as a synonym for black carbon. However, in measurement literature the terms refer to carbonaceous fractions measured with different measurement principles.

Carbonaceous aerosol (black carbon, BC and organic carbon, OC) affect the earth's radiative forcing balance and contribute to the heating or cooling of the atmosphere. Especially BC has recently received attention, since emission reductions of BC-rich sources could offer fast responses in radiative forcing (RF) but also provide health benefits (UNEP/WMO 2011, Shindell et al. 2012).

The main anthropogenic sources of carbonaceous aerosols globally are traffic, mainly from diesel vehicles, and small scale combustion (UNEP/WMO 2011, Shindell et al. 2012). Vehicle emissions are expected to decrease through new abatement technologies, for example diesel particulate filters. Small scale combustion of wood is one of the major BC emitting sources in Scandinavia and many other European countries but it does not have as strong potential for emission reductions as traffic. It is also a source where the net RF effect and public health interests might conflict.

It is essential for the air quality managers to know the major source sectors of black carbon in order to assess its' importance as an air pollutant as well as to design integrated air pollution and climate mitigation strategies. Emission inventories are an important tool in providing knowledge to support such work. This report documents the development of a black and organic carbon emission inventory for priority emission sectors for the Helsinki Metropolitan Area (i.e. the cities of Helsinki, Espoo, Vantaa and Kauniainen).

This research was implemented as a part of the WP 4.5.2 (Advanced methods to study fine particle chemistry in combustion) of the MMEA (Measurement, Monitoring and Environmental Efficiency Assessment) program coordinated by CLEEN Ltd. and funded by Tekes. The work was ordered from the Nordic Envicon Oy (Ph.D. Kaarle Kupiainen conducted the work) by Helsinki Region Environmental Services Authority (HSY). The emission activity data was mainly collected by the HSY. The novel BC and OC emission results from other MMEA research activities (MMEA WP4.5.2, 2012) were also utilized in the inventory.

2. Methodology

The basis of the black carbon emission inventory presented in this report is the already existing emission inventory of other pollutants (HSY 2011). The approach is to use the existing PM emission inventory as a basis and create BC and OC emission profiles separately for the individual subsectors utilizing the original data sources. The data in the air pollutant inventory was reviewed and it was decided that the BC inventory would at this stage be developed for priority sectors only, which are road transport, residential wood combustion and energy production. For other important sectors, for example off-road transport sources and ship emission in harbors as well as other harbor operations, further work on developing or gaining access to detailed enough background data is still required.

In case of energy production the inventory calculates plant and boiler specific emissions. In this work boiler specific BC and OC fractions were assessed and applied to the PM emissions from the inventory directly in order to derive boiler specific BC and OC emissions. The boiler specific results were further added together to get a total emission from energy production in the Helsinki Metropolitan Area.

The sector specific emissions from residential wood combustion and road transport are calculated following Eq. 1:

$$E_{i,t,p} = \sum A_{i,t} \times \left(EF_{i,t} \times f_{i,t,p}\right) \qquad \text{(Eq. 1)}$$

Where E is emissions, i is sector, t stove or vehicle category and p pollutant (BC or OC). A refers to the sector and technology category specific activity (PJ of wood or vehicle-km for on-road traffic). EF is the sector and technology category specific emission factor of particulate matter and f is the sector, technology category and pollutant specific fraction of the emission factor.

The VTT emission inventory (Mäkelä & Auvinen 2011) forms the basis for calculation of air pollutant emissions from road transport in the Helsinki Metropolitan Area. In this work the emission calculation was extended to include BC and OC profiles for the vehicle categories specified in Mäkelä & Auvinen (2011). The vehicle category specific pollutant fractions (term f in Eq. 1) were derived based on Euro-level specific information reported by Ntziachristos & Samaras (2012) combined with activity share of the different Euro-levels in Finland as in Mäkelä & Auvinen (2011).

HSY recently conducted a survey of the stove categories and fuelwood use in the residential sector in the Helsinki Metropolitan Area. With some suggested modifications the survey forms the basis for the residential sector BC and OC emission inventory. The residential sector emissions include a preliminary estimate of the potential influence of poor combustion practices on emissions.

HSY is also compiling, in collaboration with the energy companies in the area, plant specific information about PM emissions in energy production sector. In a recent research project BC emissions in selected power plants were measured (MMEA WP4.5.2, 2012) and these measurement results have been used in developing the BC emission estimates in this report.

Often the term elemental carbon is used as a synonym for black carbon although in measurement literature they refer to carbonaceous fractions measured with different measurement principles. There are several methods to estimate the amount of carbonaceous particles in aerosol samples. Some are based on light absorption of the sample (black carbon) and other popular methods determine the amount based thermal principles (elemental carbon). Differences up to 30 percent can be expected between the different measurement approaches.

Ideally an emission inventory should rely on one measurement method when gathering emission factor data. For example if the interest is to study the climate relevance of emissions then the methods measuring light absorption would be the priority. However, currently the measurement literature is too scarce to make such restrictions to the data sources. There are also other factors, e.g. combustion efficiency, combustion technologies, different fuel properties and user practices that add to the variability in measurement results making it difficult to assess the influence of measurement methodologies to individual results. For the time being it is in practice impossible to rely on results obtained with only one method. This adds to the uncertainty of the emission inventory results. This report uses for consistency the term BC. The main measurement method is reported with the emission factor data.

2.1. Road transport

Vehicle engine and emission after treatment technologies as well as fuel type are important factors affecting the characteristics of the PM exhaust emissions from vehicle traffic. BC and organic matter (e.g. OC x 1.3) form about 90 percent of the PM emissions from vehicle exhaust.

VTT has developed a comprehensive calculation system for emissions from transport sources that operates on a municipal level (see Mäkelä & Auvinen 2011). This data is used also for estimating the PM emissions in the Helsinki Metropolitan Area (HSY 2011). However, for the time being the VTT emission calculation system does not include BC and OC and therefore outside sources were used for developing the technology and fuel specific emission shares for these pollutants.

The calculation scheme was designed to follow the VTT approach and includes gasoline and diesel passanger cars, vans and buses as well as trucks with and without a trailer. The mileage split to shares of different after treatment technologies (Euro-standards) are calculated based on data given in Mäkelä and Auvinen (2011). The GAINS model databases for Finland were used for creating the technology specific emission profiles of the carbonaceous species. The carbonaceous shares are based on EMEP/EEA emissions inventory guidebook – 2009 (see Ntziachristos & Samaras, 2012). Ntziachristos & Samaras (2012) do not give details about the measurement method but based on the terminology used in the report the data has been most probably determined based on thermal optical methods. Table 1 compiles the BC and OC percentages for the Euro-level technologies in different vehicle categories used in the calculation.

Table 1. BC and OC shares in PM by Euro-level and vehicle category (Source: GAINS model).

BC share (% PM)	Cars and light trucks, gasoline	Cars and light trucks, diesel	Heavy duty traffic
Pre-Euro	19 %	55 %	50 %
Euro1/I	25 %	70 %	65 %
Euro2/II	25 %	80 %	65 %
Euro3/III	15 %	85 %	70 %
Euro4/IV	15 %	87 %	75 %
Euro5/V	18 %	55 %	75 %
OC share (% PM)	Cars and light trucks, gasoline	Cars and light trucks, diesel	Heavy duty traffic
Pre-Euro	62 %	30 %	31 %
Euro1/I	48 %	22 %	20 %
Euro2/II	48 %	14 %	20 %
Euro3/III	35 %	10 %	16 %
Euro4/IV	35 %	9 %	14 %
Euro5/V	48 %	29 %	14 %

The emissions shares in Table 1 reflect the general pattern of emission characteristics from the different vehicle categories. Diesel vehicles have a higher BC to OC ratio, usually well over one, compared with gasoline vehicles, which have a BC to OC ratios less than one. There are also technology specific differences in the emission profiles and for diesel vehicles the tendency is towards higher BC shares with more advanced Euro-stages. As a consequence of the Euro 5 limit value for PM particulate filters were introduced to cars in order to achieve the further reduction in the PM emission requirement and its profile seems to differ from Euro 4. However, the data of especially the more recent Euro stages should be reviewed when more measurements become available.

For the emission calculation the data in Table 1 was weighted with information about Euro-level specific activity shares in different vehicle categories as reported by Mäkelä & Auvinen (2011) in order to derive the vehicle category specific BC and OC fractions. These fractions were further applied to vehicle category specific emission factors and activities to calculate the emissions following Eq. 1.

It should be noted here that the emissions shares presented in Table 1 are measured in laboratory conditions, typically using a dynamometer. Compared to the measurements conducted at the roadside and within traffic in Helsinki area (MMEA 4.5.2), OC shares in Table 1 seem to be lower. The reasons for that is probably in the portion of semi-volatile hydrocarbons that are not measured in dynamometer but constitute a major fraction of organic matter in ambient measurements. These primary semi-volatile components also oxidize easily forming secondary organic aerosol that can even double the amount of organic matter related to the traffic emissions (Robinson et al., 2007).

2.2. Energy production in combustion plants (2-506 MW)

In combustion plants the fuel is burned in optimized and efficient combustion conditions and therefore they are not expected to be a major source of BC and OC. Recent measurements conducted in the MMEA project verify this expectation, since they indicated only trace amounts of carbonaceous species in the PM emissions (Table 2). The results are in line with earlier measurement data (see compilation in Kupiainen & Klimont, 2004).

Table 2. Emission shares of BC and OC in PM in power plants.

	BC (% in PM)	OC (% in PM)	Source
Hard coal	1.6 %	not measured*	MMEA WP4.5.2 (2012)
Heavy fuel oil (large boilers)	0.5 %	0.9 %	MMEA WP4.5.2 (2012)
Heavy fuel oil (small boilers)	2.4 %	0.4 %	Sippula et al. (2009)

* 1.5% is used based on 0-3% range based on a literature survey reported in Kupiainen & Klimont (2007).

The recent measurement results for hard coal and heavy fuel oil burned in large power plants (MMEA project) as well as results from Sippula et al. (2009) for heavy fuel oil burned in smaller installations (12 MW) were used as proxies and applied for the individual plants. The main fuel and the size of the boiler determined which BC emission profile was used. The direct measurement results were used for the measured boilers, but for others average emission shares were used. The PM inventory assumes no emissions from natural gas boilers and therefore no BC and OC emissions were estimated for those. The average emission shares of BC and OC in PM are shown in Table 2. The BC emissions from large power plants were determined based on an optical method (MAAP), but for smaller installations a thermal optical method was used.

It is important to note that the shares of BC and OC are connected to those of other pollutants, for example sulphate and ash that are formed from the fuel sulphur and ash components. If in the future the fuel properties will change, the emissions shares in Table 2 should be reviewed.

2.3. Residential wood combustion

HSY has recently conducted a survey of residential combustion technologies and wood use amounts in the Helsinki Metropolitan Area (Gröndahl et al., 2011). The survey included a literature review of emission factors of multiple pollutants and it has been used as a basis to estimate emissions from residential wood use in the area. The inventory included also preliminary estimation of emissions of BC and OC. The task for the work was to review the earlier assumptions about technology specific carbonaceous emission profiles as well as comment the assumptions on operator practice influences.

The PM emission factors used in the earlier inventory were left as they are. They were assessed to reflect well enough the characteristic emissions of the technologies in the HSY inventory. However, the earlier BC and OC shares were general and do not fully reflect the changes in carbonaceous emission profiles following differences in combustion efficiency. Therefore new shares for BC and OC are proposed in this work.

Residential combustion especially in Finnish stoves has unique technological and use characteristics (Tissari et al. 2007, Tissari 2008). Therefore it is important that the data used reflects these well enough. This study relies solely on Finnish measurement data for developing the BC and OC emission profiles for stoves. In case of boilers there are no Finnish measurements of BC and OC available, and Swedish data sources were used as a basis (Todorovic et al. 2007 and Boman et al., 2008). Tissari (2008) has used thermal optical methods in determining the carbonaceous fractions. Boman et al. (2008) reported a total carbonaceous fraction in boiler emissions, which was further split into BC and OC based on data from the Finnish Regional Emission Scenario (FRES) Model of the Finnish Environment Institute (SYKE).

In developing the new BC and OC shares the mass balance against PM_1 emission factor was checked. The emission fractions of other species than carbonaceous PM were taken from the original studies (Tissari 2008 and Boman et al., 2008). OM to OC ratio was assumed to be 1.8 following Tissari et al. (2007). The old and new shares are shown in Table 3.

Table 3. BC and OC shares (%) in $PM_{2.5}$ emissions in the technology categories of the HSY inventory.

	New shares		Old shares	
	BC (%)	OC (%)	BC (%)	OC (%)
	Conventional combustion			
Heat accumulating stove	40 %	19 %	40 %	20 %
Sauna stove	41 %	31 %	40 %	20 %
Wood boiler (with accumulator)	28 %	19 %	10 %	10 %
	Poor combustion			
Heat accumulating stove	28 %	38 %	40 %	20 %
Sauna stove	20 %	42 %	40 %	20 %
Wood boiler (without accumulator)	15 %	42 %	10 %	10 %

The emission shares for heat accumulating stoves (masonry heaters, varaava takka) for both normal and bad combustion practices are based on the emission profiles presented in Tissari (2008). For sauna stoves (puukiuas) the BC and OC emission shares for normal combustion are also from Tissari (2008). Measurement data suggests that when the combustion efficiency becomes less, OC is most responsible of the increase in the total emissions. This can be seen when comparing the normal and poor combustion practice emission profiles in Tissari (2008) in case of masonry ovens. Since there were no direct measurements available for smoldering combustion in sauna stoves, following assumptions were used to derive the relevant profile: (1) BC emission factor remains the same as for normal combustion practice, (2) OC is responsible for the increase in total PM emission factor. Figure 1 shows the speciation of the technology specific PM_1 emission factors for both normal and poor combustion practices.

Figure 1. Speciation of technology specific PM_1 emission factors for normal and poor combustion practices. OC was converted to OM applying a factor of 1.8 following Tissari et al. (2007).

The earlier inventory assumed 50/50 wood use shares for the normal and poor combustion practices, respectively. Although there are no surveys available from Finland, the assumption for poor combustion practice seems relatively high, especially taking into account the emission characteristics assumed for the practice. Therefore another major proposed change compared with the earlier inventory is in the share of wood use in bad combustion practice. The emissions presented in this report are calculated using the revised combustion practice shares.

Finnish Regional Emission Scenario model (FRES) managed at the Finnish Environment Insistute (SYKE) provides the Finnish national BC and OC emission estimates (see for example Arctic Council 2011). In case of stoves the average emission profile of the FRES model assumes that approximately 10 percent of stove fuel use is done following the poor combustion practice. That order of magnitude is similar to the results from Norwegian and Swedish surveys (Haakonsen & Kvingedal 2001, Todorovic et al. 2007), although the technologies are not fully comparable with the Finnish combustion.

Share of wood use in boilers with and without heat accumulator is assumed 50/50 in the HSY inventory, respectively. Based on a recent Finnish national survey by Metla (Torvelainen 2009) this seems an overestimate. For the whole country Metla assessed that approximately 20 percent of boiler fuel use takes place in boilers without a heat accumulator. This share is also recommended to be used by HSY until better data becomes available. The emissions presented in this report are calculated using the revised share.

2.4. Other

Due to data constraints emissions from the potentially important BC source sector, off-road transport (including emissions from ships in harbors), are not calculated in this work. Developing suitable data should be seen as a priority for further development of the inventory.

3. Results and discussion

Figure 2 presents the total emissions of BC and OC in 2010 in the Helsinki Metropolitan Area for the estimated sectors. Total emissions of BC and OC are estimated at 235 and 70 tons in 2010, respectively. On-road transport is the most important source sector comprising 83 and 58 percent of the BC and OC emissions, respectively. Practically the rest of the total emissions are emitted from residential combustion of wood. Energy production in large combustion plants is only of minor importance and accounts for approximately 1 percent of emissions of BC in the area.

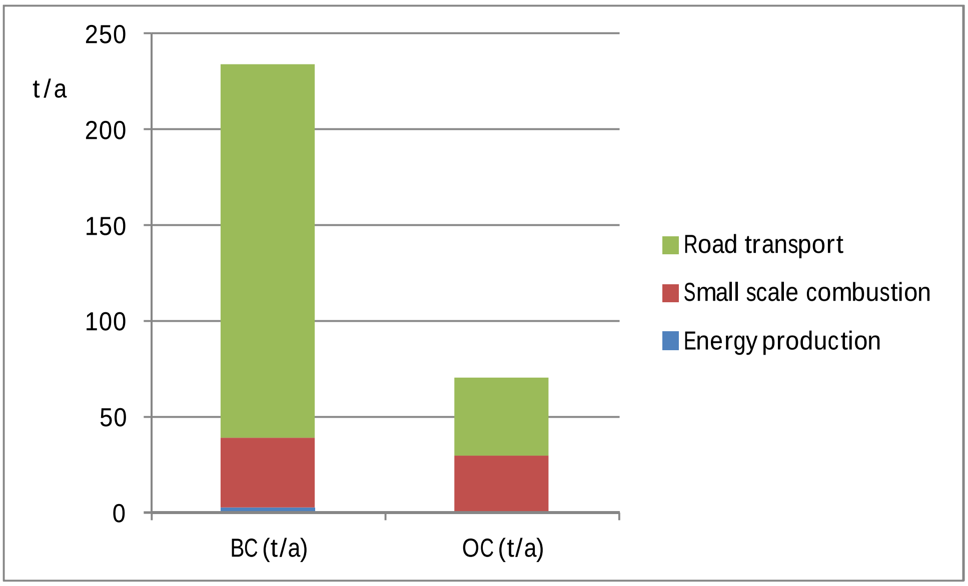

Figure 2. Emissions of BC and OC in the Helsinki Metropolitan Area in 2010.

A more detailed split of the BC emission from the on-road transport sector by vehicle category is presented in Figure 3. Diesel vehicles are responsible for the BC emissions almost completely. Diesel passenger cars are responsible for 40 to 50 percent; diesel vans about 25 percent and diesel heavy duty vehicles approximately 30 percent of the total emissions in each city.

The contributions from wood burned according to normal and poor combustion practices to BC and OC emissions are presented in Figure 5. The assumed portion of wood burned with poor combustion practice was 10 percent for stoves and 20 percent for boilers (see Section 2.3). The fuel use estimates were combined with emission factors presented in Figure 1).

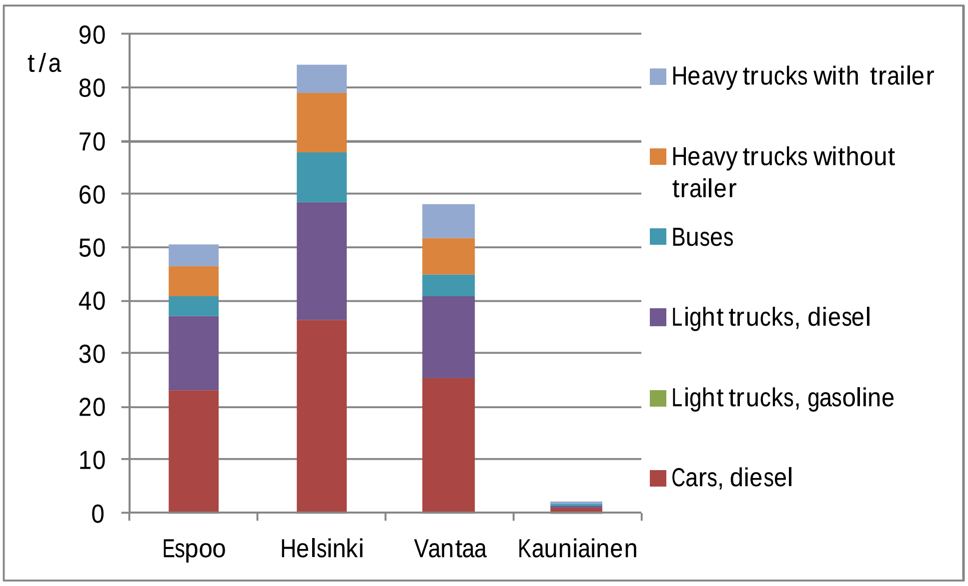

Figure 3. BC emissions in 2010 from the on-road transport sector by vehicle category in the studied cities.

Figure 4 shows the emissions from residential combustion split into the three stove categories. Heat accumulating stoves (varaavat takat) account for 65 and 59 percent of the BC and OC, respectively, and are followed by sauna stoves that account for approximately third of the emissions. Emissions from boilers comprise 3 and 6 percent of BC and OC, respectively.

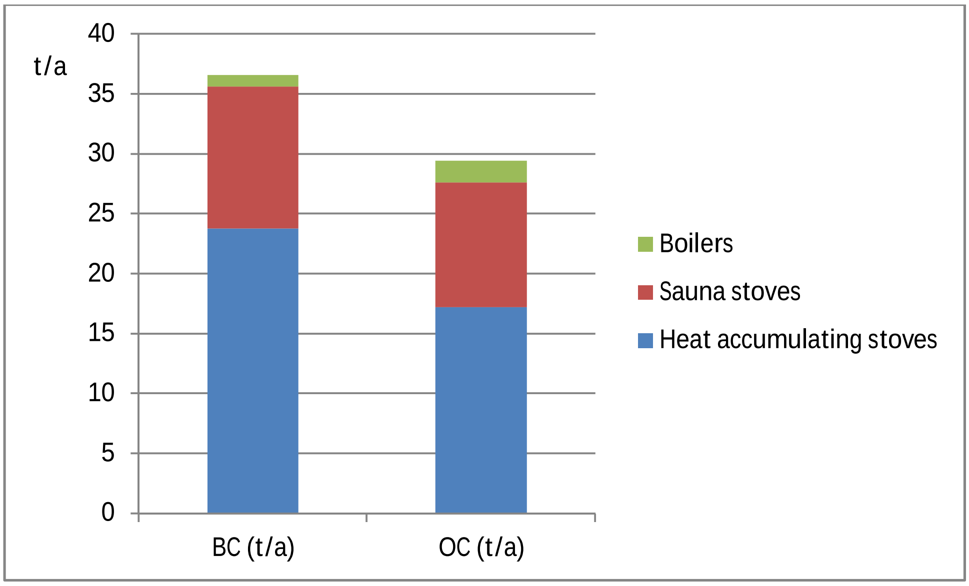

Figure 4. BC and OC emissions from residential combustion by stove category.

Results in Figure 5 show that emissions from poor combustion have a significant share in all of the emission categories but are more pronounced in the OC emissions. In order to estimate the potential to reduce emissions of BC and OC by influencing the user practices in the residential sector the results in Figure 5 should be interpreted together with data presented in Figure 1. In case of sauna stoves there is only limited potential to reduce BC emissions, because the emission profiles of normal and poor combustion practice have practically the same emission factor. In all other cases switching from poor to normal combustion practice could result in significant emission reductions in both BC and OC.

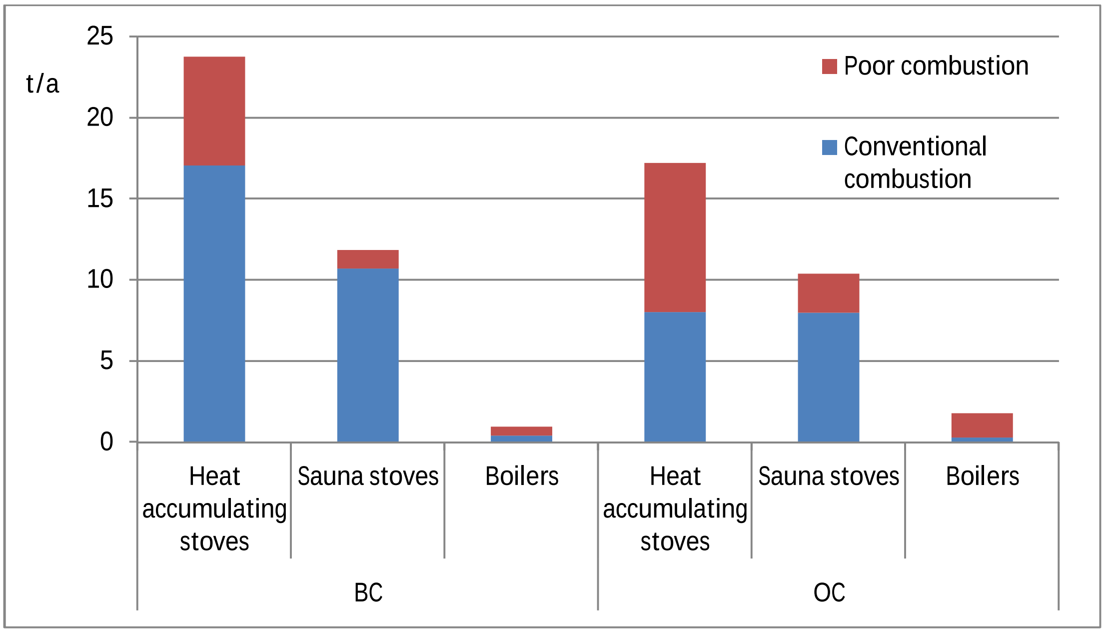

Figure 5. BC and OC emissions from residential combustion by technology and combustion type.

4. Conclusions and recommendations

Based on the emission inventory of BC and OC, transport and residential wood combustion are the most important sources of carbonaceous particles in the Helsinki Metropolitan Area in 2010. Applying the very recent measurement results shows that large energy plants are only of minor importance when emissions of carbonaceous particles are under focus. This is expected because the combustion occurs in optimized conditions.

The current carbonaceous emission estimate is not yet comprehensive in covering all relevant sectors. A potentially major emission source sector, off-road transport, is missing completely from the current estimate due to lack of background data. Including it into the inventory should be seen as a priority in developing future work plans.

Residential combustion emission calculation for the Helsinki Metropolitan Area has a rather coarse technological structure compared for example with another available Finnish BC inventory, as well as the data available from the surveys conducted in the Metropolitan Area. The Finnish Regional Emission Scenario model (FRES) managed at the Finnish Environment Institute (SYKE) has currently more technological subcategories in the residential sector than the HSY inventory and it is recommended that HSY studies the possibility to develop their calculation schema towards similar technological detail and parameterization as in the FRES model in collaboration with SYKE.

This report discusses emission of primary particulate BC and OC. In ambient measurements a substantial proportion of OC is formed from gas-phase organic compounds (VOCs) which transfer to particle phase after emission via oxidation reactions in the atmosphere.

5. Literature

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6. Appendix: Emissions (tons/year) of PM, BC and OC in the Helsinki Metropolitan Area in 2010.

	PM	BC	OC
Residential wood combustion			
Heat accumulating stove	67	24	17
Sauna stove	32	12	10
Wood boiler	5	1	2
Subtotal	104	37	29
Road transport			
Cars, gasoline	5	1	2
Cars, diesel	109	85	15
Light trucks, gasoline	0	0	0
Light trucks, diesel	67	52	10
Buses	25	17	5
Heavy trucks without trailer	35	24	6
Heavy trucks with trailer	22	15	4
Subtotal	262	194	41
Energy production	158	2	1
TOTAL	524	233	72