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Primary particulate matter emissions and the Finnish climate strategy

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There is an increasing need for the assessment of particulate matter (PM) emissions of different particle sizes because of the adverse health effects of the fine size fractions. Combustion of fuels is an important source of fine primary particles and an important factor in strategies to combat climate change. In this study, a calculation system was developed for primary PM emissions from stationary combustion and their control. It was used to calculate present (1990 and 1995) and future (2010) primary PM emissions for three energy scenarios. Large energy production units already utilize very efficient particle removal equipment. Less extensive use of solid fossil fuels in studied low-CO_2 scenarios does not substantially reduce primary PM emissions. Domestic wood combustion, where no control technologies are in use, constitutes the major part of combustion-based primary PM emissions both at present and in the future, especially for the fine particle fractions. The results for 1995 were considerably higher than earlier international estimates.

Introduction

There is a growing concern about the health effects of fine particles. Recent studies have demonstrated consistent associations between concentrations of particulate matter (PM) and adverse effects on human health (respiratory symptoms, morbidity and mortality) at concentrations commonly encountered in Europe and North America. Consequences for public health may be considerable (e.g. Künzli *et al*. 2000). Integrated assessment models have been used to identify least-cost strategies to control multiple precursor emissions leading to acidification, eutrophication and ground-level ozone at international (e.g. Amann *et al*. 1999) and national level (e.g. Johansson *et al*. 2001). The European version of the RAINS (**R**egional **A**cidification **IN**formation and **S**imulation) model has recently been extended to address control strategies for different size fractions of PM (Amann *et al*. 2001, Lükewille *et al*. 2001). This model framework, which will be used as a tool in future negotiations on European emission reductions, will utilise data on national emission characteristics provided by sources such as this study.

Airborne suspended PM can be either primary or secondary in nature. Natural and anthropogenic processes emit primary particles directly into the atmosphere. Secondary particles have predominantly man-made origin, since they are formed in the atmosphere from oxidation and subsequent reactions of sulphur dioxide, nitrogen oxides, ammonia and volatile organic compounds.

PM concentrations in Finland are affected both by local and transboundary atmospheric transport. Both primary and secondary particles and their precursor gases can travel long distances, especially the fine fraction (PM2.5, aerodynamic particle diameter < 2.5 μm). Particle concentrations in urban areas are typically dominated by local emissions, mainly from traffic. Many single-family houses in residential areas are equipped with wood-fired main or supplemental heating devices and sauna stoves with relatively low stack heights and no emission control devices. Thus primary particles from wood combustion may locally contribute to remarkable concentrations of fine particles, especially in cold winter days when heating devices are intensively used and atmospheric mixing is poor due to an inversion. Outside central urban areas transboundary impacts are relatively more important. Secondary particles, which are mainly of transboundary origin, are large contributors to fine particle concentrations in rural background areas (EMEP/MSC-W 2000).

The concentration levels of total suspended particles (TSP) and PM10 in Finland are in general high enough to be of concern. National daily and annual guidelines and limits for TSP and PM10 have been exceeded in a large share of measurement stations, mainly in urban areas during the 1990s (Kukkonen *et al.* 1999, Pietarila *et al.* 2001). Annual concentrations of PM10 in several downtown areas have varied between 20 and 40 μg m^{-3} (Pietarila *et al.* 2001). The levels of PM10 and PM2.5 have been about 20 μg m^{-3} and 8–12 μg m^{-3}, respectively, in the Helsinki metropolitan area in the late 1990s (Johansson *et al.* 2002). Experimental studies have helped to associate ambient air quality with personal exposures (Koistinen *et al.* 2001) and risks for health effects (Pönkä *et al.* 1998). Inorganic secondary particles were found to be the major contributor of personal exposures on PM2.5 in Helsinki, while the estimated share related to combustion was roughly one third of the exposure (Koistinen *et al.* 2003).

The main characteristic of the Finnish economy relevant to air emissions is a large-scale exploitation of forests, leading to both high energy needs because of energy-intensive forest industries, and the extensive use of different kinds of wood fuels. The most important primary energy carriers are fuel oils (27% of the total primary energy consumption of 1308 PJ in 2000), wood fuels (21%), nuclear (18%), natural gas (11%) and coal (8%) (Statistics Finland 2001). Black liquor combustion in the paper pulp industry is the most widely used wood fuel (53% of the total wood fuel use). The other sources are combustion of forest residues in industry and power plants (30%) and domestic small-scale wood combustion (17%). The main anthropogenic emission sources of primary particles are fuel combustion, industrial processes and fugitive dust sources. Natural sources include, e.g., wind erosion of soils and sea salt. Mechanically produced dust particles are generally quite large in size. Primary fine particles originate mainly from the combustion in stationary and traffic sources and from industrial processes.

In this study, a calculation system was developed for primary PM emissions and their control in Finland from stationary combustion sources, i.e. energy production in power plants and industry, and small-scale combustion in the domestic sector. Primary emissions of TSP, PM10 and PM2.5 were calculated for 1990 and 1995, and for 2010 using three national energy scenarios of the Finnish climate strategy: ‘Baseline’, ‘Kyoto-gas’ and ‘Kyoto-nuclear’ scenario. The primary PM emission calculations for 2010 formed part of the environmental impact assessment of the Finnish climate strategy (Hildén *et al.* 2001). The primary PM emissions from traffic exhaust and industrial processes were also roughly estimated in that assessment study (Syri *et al.* 2001).

Primary emissions of TSP have been annually reported in Finnish emission inventories for stationary and traffic combustion sources, as well as industrial processes (Statistics Finland 2001). Finnish fine particle emission factors have been estimated for some sectors of large-scale energy production (Ohlström *et al.* 2000). In this study, the first quantitative estimates on the fine primary PM emissions from stationary combustion in Finland are calculated. This study also provided data on the PM10 and PM2.5 fractions of TSP for stationary combustion in the officially reported Finnish emission inventory to EMEP for 2000. The results are compared with data from a Europe-wide primary PM emission inventory for 1995 of CEPMEIP (the Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance), which aims to support national experts in the official reporting of primary PM emission inventories (EMEP/MSC-W 2002).

Methods and data sources

Different combustion processes form particles of different sizes and chemical composition. Solid fuel combustion in power plants and large industrial boilers produces mainly ash particles, formed from non-combustible mineral matter, the fraction of carbonaceous particles being generally less than 10% (Lighty *et al.* 2000). Mineral matter forms mainly relatively large particles (1–50 μm). Only a small part of the mineral matter, usually less than 10% (Lind *et al.* 1996), volatilises in high temperatures. The volatilised compounds nucleate or condensate forming ultrafine particles (< 0.1 μm). The ultrafine particles grow into accumulation mode (0.1–1 μm) by coagulation and condensation. Combustion of oils, rarely containing high fractions of mineral species, produces mainly supermicrometer coke particles and ultrafine soot particles with organic compounds condensed on the surfaces of the particles (Flagan and Seinfeld 1988). Carbonaceous soot particles are formed from volatilised hydrocarbons that nucleate and grow via chemical surface reactions and coagulation. Particles from small-scale wood combustion in the domestic sector contain high fractions of fine soot and condensed organic compounds (Ålander 2000, Rogge *et al.* 1998, Rau 1989), and emission factors may be relatively high, since emission controls are not utilized and combustion processes are in most cases poorly controlled (McDonald *et al.* 2000, Purvis *et al.* 2000, Hahkala *et al.* 1986). PM emissions in appliances with well-controlled combustion conditions, e.g. pellet boilers, can be an order of magnitude lower than in conventional stoves (Tullin *et al.* 2001). In power plants and industry the major part of the particles is currently removed by emission control appliances, such as electrostatic precipitators (ESP). ESPs remove particles below 3 to 5 μm considerably less efficiently than coarser particles (Ylätalo and Hautanen 1998, McElroy *et al.* 1982).

In this study, Finnish primary PM emissions, e_{m}, were calculated from activity data a (i.e. primary energy consumption, PJ a^{-1}), unabated emission factors, e_{f} (i.e. emission factors without emission control devices, mg MJ^{-1}), and emission removal efficiencies, η (fraction), of various emission control technologies (5 different technology options) which can be applied to each sector-fuel type combination (6 sectors, 11 fuels) with certain utilization rates, x (fraction). The PM emission for time t, emission (TSP, PM10 or PM2.5) e, fuel i, sector or combustion technology j and and control technology k is

$$e_{\mathrm{m}}^{e}(t) = \sum_{i}\sum_{j}\sum_{k}\left(1-\eta_{i,j,k}\right)x_{i,j,k}(t)a_{i,j}(t)e^{e}{}_{i,j} \quad (1)$$

The unabated emission factors of PM10 and PM2.5 were derived from unabated TSP emission factors using estimated fractions of PM10 and PM2.5 from total particle mass, y^{PM10} and $y^{\mathrm{PM2.5}}$, respectively, and assuming that $y^{\mathrm{TSP}} = 1$,

$$e_{\mathrm{f}}^{e} = y_{i,j}^{e}\, e_{\mathrm{f}\,i,j}^{\mathrm{TSP}} \quad (2)$$

The fuel, sector and combustion technology specific unabated emission factors and control technology removal efficiencies for different particle size classes were based on particle size distributions of national and other measurement studies in the literature (e.g. Johansson 2002, Ohlström *et al.* 2000, Mikkanen *et al.* 1999, Moisio 1999, Ylätalo and Hautanen 1998, Lammi *et al.* 1993, Hahkala *et al.* 1986). All data sources used are presented in more detail in Karvosenoja (2001). The utilization rates of emission control technologies of the existing energy system were determined based on data of the register on air pollution permits of the Finnish environment administration VAHTI (Korkia-Aho *et al.* 1995) containing technical and emission information on the Finnish industrial and energy producing plants exceeding a thermal capacity of 5 MW$_{\mathrm{th}}$. In the scenarios, the utilization of emission controls in new capacities was estimated based on legislative requirements, e.g. the Large Combustion Plants Directive of the European Union (EU 2001).

The activity data for 1990 and 1995 were determined using national statistics (Statistics Finland 2001). The activity data in 2010 were based on three national energy scenarios of the Finnish climate strategy by the Ministry of Trade and Industry (2001). The Technical Research Centre of Finland has compiled the scenarios with the energy system model EFOM-ENV (e.g. Lehtilä and Tuhkanen 1999). All scenarios assume an average annual economic growth of 2.4%. The first scenario 'Baseline' does not include any additional restrictions on energy production system, leading to a 16% increase in the total primary energy consumption in 2010 compared to 2000. This increase would mainly be met by a more extensive use of hard coal, and the greenhouse gas (GHG) emissions would increase by about 20% from 1990. The EU burden sharing agreement for Finland is the stabilization of emissions at the 1990 level. In the other two scenarios, 'Kyoto-gas' and 'Kyoto-nuclear', this agreement would be met. Both Kyoto-scenarios include GHG emission reduction measures, e.g. more emphasis on energy saving and fuel switching to low carbon content fuels in centralized heat and power production. 'Kyoto-gas' includes a strong shift from coal to natural gas and biomass. In Kyoto-nuclear, one new 1400 MW_e nuclear reactor would be introduced to the Finnish energy system, with a moderate shift from coal to biomass and gas. The scenarios are presented in more detail in Hildén *et al.* (2001) and Syri *et al.* (2001).

Results

The Finnish unabated and actual emissions of TSP, PM10 and PM2.5 from stationary combustion sources were calculated for 1990, 1995 and 2010 with three scenarios (Fig. 1). Primary PM emissions are currently effectively controlled in power plants and industry. Almost all the large solid fuel combustion boilers were equipped with ESPs already in 1990. Decrease in emissions between 1990 and 1995 was mainly due to the introduction of flue gas desulphurisation (FGD) technologies in coal power plants and black liquor recovery boilers. Retrofitting the plants with FGD equipment were made in order to achieve the target set by the Finnish Second Sulphur Committee (1993) for reducing sulphur dioxide emissions by 80% by the year 2000 from the 1980 level. In 1995, 82% of coal used in power plants, and 90% of black liquor were combusted in plants equipped with FGD. The use of wet FGD, or spray dry scrubbers combined with fabric filters reduces also PM emissions considerably (Ohlström *et al.* 2000). In 1995 the average removal efficiency in Finnish power plants and industry was higher than 99% for TSP and higher than 98% for PM2.5. Black liquor combustion in paper pulp production was the major source of emissions in industry, especially for PM2.5. Although the unabated emissions from combustion of black liquor and solid fuels in power plants and industry would have accounted for 99% for TSP and 97% for PM2.5 of the total stationary combustion based emissions in 1995, the actual emissions accounted for only 40% and 30%, respectively.

The emissions from wood combustion in the domestic sector, where emission control measures are not utilized, are high, especially for PM2.5. Heating with wood is common in residential neighbourhoods and sparsely populated areas in Finland. Many old stoves and fireplaces presumably have high emission factors. Most of the well-maintained modern wood heating devices have technically well-controlled combustion conditions and thus considerably lower emissions, assuming that fuel quality is also appropriate. In 1995 domestic wood combustion emitted 14 Gg PM2.5, accounting for more than half of the total stationary combustion PM2.5 emissions. Including primary PM2.5 emissions of 1998 from combustion in traffic and industrial processes (Syri *et al.* 2001), the share for domestic wood combustion would be roughly 40% and for combustion in power plants and industry less than 25%.

In 2010 the PM emissions will remain roughly at the 1995 level for all three scenarios. Although more extensive use of coal in the Baseline scenario causes a steep increase in the unabated emissions, the actual emissions from new coal power plants, after ESP and wet FGD

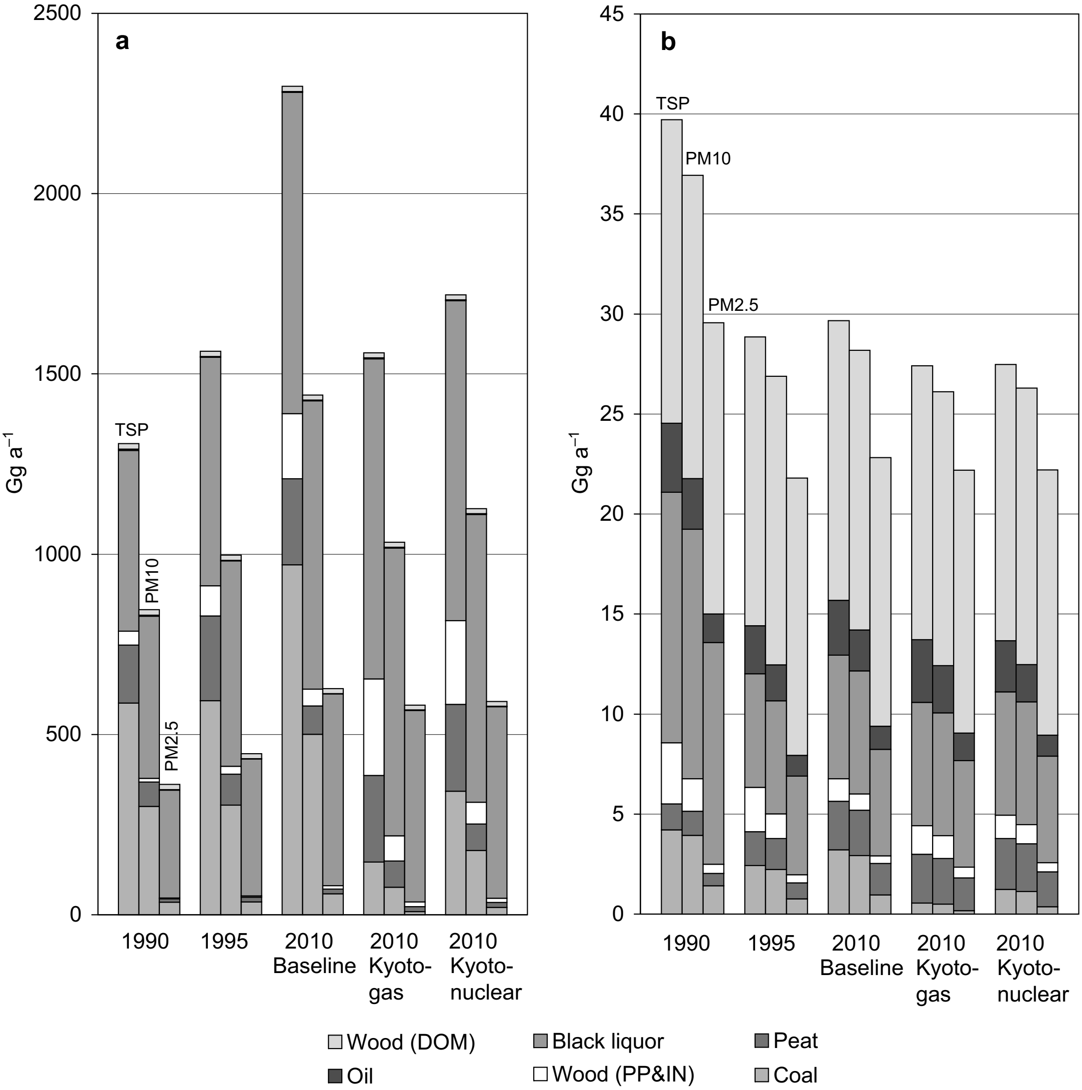

Fig. 1. The Finnish annual unabated (**a**) and actual (**b**) emissions of TSP, PM10 and PM2.5 from stationary combustion sources in 1990, 1995 and 2010 with the 'Baseline' and 'Kyoto-gas' scenarios. (DOM = domestic combustion, PP&IN = combustion in power plants and industry.)

equipment, are relatively low. Differences in PM emissions between the Kyoto-scenarios are small: slightly higher emissions from the use of oil and fuel wood in industry in 'Kyoto-gas' are compensated with the emissions caused by the higher use of old coal power plants in 'Kyoto-nuclear'. In general, the emission levels in new large combustion plants are regulated by legislation, mainly the Large Combustion Plant Directive of the EU (2001), and thus standards of emission reduction technologies are high. The amounts of wood combustion in the domestic sector and black liquor combustion in industry, which cause the major fraction of the emissions, are practically at the same level in all three scenarios. Wood combustion in the domestic sector increases slightly from 1995, but the emissions do not increase due to assumed renewal of combustion equipment, and thus decrease in average emission factors. For the traffic sector and industrial processes, differences between the scenarios are small (Syri *et al*. 2001).

Comparison with national statistics and the CEPMEIP inventory

The TSP emissions for 1990 and 1995 calculated in this study were compared with the corresponding values in the Finnish statistics (Table 1). The emission values in the statistics are based on the ILMARI model that calculates air emissions on a plant-by-plant basis from 2200 combustion and industrial processes (Statistics Finland 1998). There is a relatively good agreement for 1995 for the fuels used mainly in power plants (i.e. other than wood). The greatest difference lies in peat combustion, where the value of this study is 42% higher than in the statistics. The category 'other boilers' in the statistics refers mainly to the co-combustion of peat and wood. In the terms of both combustion technique and fuel supply, peat and wood are well suited to be used simultaneously, and they are often co-combusted in the same boiler. In this study the emissions from co-combustion of several fuels are allocated to the respective fuels. Therefore these and statistics values are not fully comparable for peat and wood.

The calculations in this study give lower emissions from power plants and industry in 1990 than the statistics. The difference is partly due to insufficient data on the utilization rates of control technologies in 1990.

The calculations of this study give lower emissions for wood combustion than the statistics both in 1990 and 1995. The statistics do not separate domestic and industrial combustion. However, for wood combustion emissions are predominantly from domestic sources. The estimates on the utilization rates of different types of small-scale wood combustion appliances have considerable uncertainties, and the PM emission factors for various stove types vary greatly. Thus the emission values for domestic wood combustion are very uncertain. The emission values in the statistics encounter the same problem of uncertainties in the domestic sector. (K. Grönfors pers. comm.)

The results of this study and the CEPMEIP inventory in 1995 for stationary combustion sources are compared in Table 2. Emission estimates by CEPMEIP are substantially lower for all the sectors than results from this study. The activity rates are roughly the same in both studies. The CEPMEIP inventory employs default emission factors, which do not reflect country-specific circumstances. The assumptions for the utilization rates of emission control technologies in power plants and industry sectors (SNAP 1 and 3) are not clear, and thus possibly constitute the main reason for the differences in these sectors. Emission factors for domestic wood combustion are clearly lower than in this study.

Discussion and conclusions

In this study, a Finnish calculation model for TSP, PM10 and PM2.5 primary emissions from stationary combustion sources was developed,

Table 1. The Finnish TSP emissions from stationary combustion sources in 1990 and 1995 based on this study and the national statistics (Statistics Finland 2001; K. Grönfors pers. comm.). The statistics do not separate emissions from domestic and industrial sources. The category 'other boilers' in the statistics refers mainly to the co-combustion of peat and wood. In this study the emissions from co-combustion of several fuels are allocated to the respective fuels. (DOM = domestic combustion, PP&IN = combustion in power plants and industry.)

	1990 this study (Gg a^{-1})	1990 stat. (Gg a^{-1})	Differ. from stat. (%)	1995 this study (Gg a^{-1})	1995 stat. (Gg a^{-1})	Differ. from stat. (%)
Coal	4.2	5.8	–27	2.4	2.3	5
Peat	1.3	4.3	–70	1.7	1.2	42
Oil	3.5	3.8	–9	2.4	2.6	–7
Black liquor	12.5	15.7	–20	5.7	5.5	3
Wood, DOM	15.2	20.0	–9	14.4	19.9	–16
Wood, PP&IN	3.1			2.2		
Other boilers	–	7.6	–	–	3.1	–
Total	39.7	57.2	–31	28.9	34.6	–17

and the effect of national energy projections on primary particulate matter emissions was studied. Combustion processes are an important source of fine PM emissions, as an emitter of both primary particles and precursor gases of secondary particles. Combustion of fossil fuels also contributes to the major part of the anthropogenic greenhouse gas emissions globally.

The results of this study and from a detailed model by Statistics Finland exhibited comparable emissions from stationary combustion. The comparison with the European-wide CEPMEIP inventory suggested that national characteristics may not be fully reflected by common default emission factors. Both comparisons highlighted the importance of using correct utilization rates of emission control technologies. In general, emission calculation models aimed to scenario analyses, such as the one developed in this study, operate with a limited number of sector and fuel type combinations. There is a high within-sector variability of e.g. emission factors when compared to plant-specific data. For long-term regional scenarios other input variables become equally or more important, such as activity levels and largely lacking measurement data for small appliances burning biomass in the domestic sector. The quantification of these factors will be the main task for this calculation model in the future.

Primary PM emissions from large energy production units are currently effectively controlled in Finland. In 1995 fuel combustion in power plants and industry accounted for roughly one third of the primary PM2.5 emissions from stationary combustion sources. Domestic wood burning, where devices are not equipped with emission controls and have relatively high emission factors in old heating devices, accounted for more than half. Stationary combustion sources contribute to more than half of the total primary PM2.5 emissions including traffic and industrial sources.

The two post-Kyoto energy scenarios, which assumed different mixes of energy sources, indicated only slightly lower primary PM emission than the baseline scenario. The choice of fuel in new large energy production units, whether using coal, natural gas or nuclear fuel, did not have a remarkable effect on primary PM emissions due to required effective emission controls.

Table 2. The Finnish PM emissions from stationary combustion sources in 1995 based on this study and the CEPMEIP inventory (data downloaded from http://www.air.sk/tno/cepmeip in September 2002). (SNAP 1 = Combustion in energy and transformation industries, SNAP 2 = Non-industrial combustion plants, SNAP 3 = Combustion in manufacturing industries.)

	TSP this study (Gg a^{-1})	TSP CEPMEIP (Gg a^{-1})	differ. from CEPMEIP (%)	PM10 this study (Gg a^{-1})	PM10 CEPMEIP (Gg a^{-1})	differ. from CEPMEIP (%)	PM2.5 this study (Gg a^{-1})	PM2.5 CEPMEIP (Gg a^{-1})	differ. from CEPMEIP (%)
SNAP 1	3.3	1.5	120	2.9	1.4	107	1.2	1.2	5
SNAP 2	15.0	9.1	65	14.9	8.0	86	14.2	7.3	95
SNAP 3	10.6	4.6	130	9.3	3.2	191	6.6	2.3	18
Total	28.9	13.7	111	27.1	12.6	115	22.0	10.8	104

Wood combustion in the domestic sector is the main contributor to primary fine PM emissions and its share is expected to increase slightly from 1995 in all three energy scenarios. The PM emissions, however, do not increase because of broader use of modern low-emission combustion equipment. The expected increase in small-scale wood combustion would result in roughly 20% increase in PM2.5 emissions of this sector and 10% increase in Finnish total primary PM2.5 emissions in 2010, if current emission factors were used. An introduction of emission standards for new stoves would help reducing emission from this sector. The increasing use of biomass in small-scale combustion has been considered a potential way to reduce greenhouse gas emissions, but possible negative side effects should be explored further.

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