**Responses to reviewers of the paper by**

**Klimont et al. (http://www.atmos-chem-phys-discuss.net/acp-2016-880/)**

**RESPONSES TO Reviewer 1:**

We thank the reviewer for a very thorough and insightful review which we have used to improve the manuscript and provide additional results in further extended Supporting Information set. The responses to specific points raised by the reviewer are provided below.

REVIEWER:
The calculations often incorporate substantial technology and emission control details, however the results of this detail are only presented at a fairly aggregate level. Some key intermediate results here would be very useful to present. In particular, for some key sector/fuel combinations, I suggest that the emission factors over time for different regions (perhaps PM or BC in the main paper, and other species in the supplement). One key sector is onroad diesel, for example, where aggregate emission factors have changed over time in many regions. Other key sectors might be residential biomass, off-road diesel, etc. Where these emission factors are largely constant, this could be mentioned in the text without a figure, but where these have changed some figures and/or tables and some discussion would be useful

RESPONSE: Indeed, the model considers explicitly implementation of particular technologies to achieve required emission standards. We produced now a specific output for selected sectors (on-road diesel, off-road diesel, power –coal, industry-coal, residential biomass) and all key regions. On the basis this output we present a few examples where changes were relevant within the characterized period.

REVIEWER:
The main presentation focuses on emissions by region, and global emissions by sector. There is a lot of material here, and this is a reasonable choice for the main paper, however many readers will want to see emissions by sector for specific regions. It would be useful to present the equivalent of Figure 3 for the different regions in the supplement. (Hopefully the codes that generated that figure can readily be generalized.). As noted below the authors should also provide an electronic supplementary file with more detailed emissions. Some further suggestions for details that would be useful to supply are below.

RESPONSE: We added now two additional outputs:

- The set of tables presenting emissions for each of the 25 IMAGE regions by key sectors (power-coal, power-other fuels, industry, coke ovens, fossil fuel production, residential – biomass, res –coal, res – other fuels, road transport –diesel, road transport – other fuels, non-road – diesel, non-road – other fuels, other). This is an Excel file that will be part of the SI.

- A set of figures for key regions (as in Figure 3) with emissions for PM species over time as in Figure 4. We believe that the reviewer meant Figure 4 for key regions rather than Figure 3 since the latter is already regional. These additional figures are in the SI.

REVIEWER:
Overall, its not clear how equipment vintages are treated. There is a mention of old/new power plants, but not as much discussion for other sectors. For vehicles, for example, is the model based on aggregate emission factors by year, or are vintages of vehicles tracked over time? This should be clarified in the manuscript.

RESPONSE: The equipment vintages are dealt in GAINS in two ways. Explicit assumptions for power sector where activity (energy use) for existing and new (post 2000) power plants are included, and specifying pace of equipment replacement, using technical lifetime of technologies in GAINS database which is defined for each control technology, in the so called 'control strategies' where share of fuel use for a given technology is given. While for many sectors, the add-on control technologies can be applied at any time and vintage plays smaller role, for transport new standards most of the time are synonymous with a new vintage year of a particular vehicle. Vehicle lifetimes are region specific and affect the fleet turnover which in turn determines how quickly a new technology penetrates the market. As for vehicle emission factors, in GAINS they represent for each of the categories, e.g., EURO 1, EURO 2 or any other standard, the average lifetime emission rate, including typical deterioration factors. We add additional text in the manuscript to explain how vintages of installations and technologies are dealt with – see section 2.2.2 and 2.2.3.

REVIEWER:
Specific comments Section: 2.2.1 Residential combustion: cooking, heating, lighting: It is not clear if assumptions such as the use fraction of different technologies and also splits between end-use services (e.g. cooking, heating in particular) are constant over time within a given country or region?

RESPONSE: There is an explicit region-specific assumption in the model about the fuel use for cooking and heating. For Europe and North America solid fuels in residential sector are allocated to heating unless specific information exist, e.g., Switzerland provided their estimates for cooking. We are aware that there is some cooking (or cooking and heating using the same devices) in several countries but there is no data and the cooking share is most likely very small compared to demand for heating. For Asia, Africa, and Latin America cooking dominates although for some countries like China and few of the Latin American countries heating plays an important role; similarly in some provinces (states) in India, Pakistan, or Nepal. The data for that originates from the GAINS-Asia related studies as well as the recent work in Latin America with support of CCAC and UNEP (final report in preparation for publication; see summary for policy makers http://www.ccacoalition.org/en/resources/integrated-assessment-short-lived-climate-pollutants-latin-america-and-caribbean-summary). The share of cooking/heating is assumed constant in the 1990-2010 period as we have not found any data allowing to change that assumption. The manuscript has been modified to reflect the above discussion by adding additional references (see 2nd paragraph of section 2.2.1) and a specific statement about the constant ratio of cooking/heating (see the last sentence in section 2.2.1 which reads "*The ratio of cooking/heating is assumed constant in the 1990-2010 period as we have not found any data allowing to change that assumption*"

As far as technology split is concerned; there is no firm statistical data but for several countries our exchange with national experts led to adjustment of assumptions how the technology (stoves/boilers/automatic boilers/pellet stoves/fireplaces) shares changed over time. Again, there is no model behind but an attempt to reflect on available information from local studies or expert contacts. For example sales statistics, for example for pellet stoves and boilers, resulted in adjustment of shares of biomass used in such installations in several European countries where strong growth has been observed towards the end of the period under investigation and continues into the future (another paper). Also for China, trends towards cleaner coal stoves and more household coal boilers (in specific provinces) were taken into account. The manuscript has been modified including explicit statement that the technology shares change over time and addition of new references (see 1st paragraph in section 2.2.1).

Finally, residential use of kerosene was split into cooking and lighting and the shares change over time depending on the regional access to electricity, as described in the manuscript (see section 3.2).

REVIEWER:
Specific comments Section: 2.2.1 Residential combustion: cooking, heating, lighting: There is quite a bit of good work here and some sort of summary later on in terms of the evolution of aggregate emission factors (either over time – if the above assumptions change over the historical period) would be useful. For example, do PM emissions per unit biomass fuel use for cooking change over time in a given region? (Some of this could be in the supplement, with main points summarized in the text). While there are too many details to do this comprehensively, it would be important to summarize where emission factors for important sectors change over time (either as a result of different technology fractions, or emission controls). This is clearly going to be the case for diesel fuel use in road transportation in many regions, but how much change was assumed for the various regions. How about off road, agriculture, or residential sectors? Did controls or technology mix (beyond shifts in type of fuel use) have a noticeable impact on emission factors in these regions?

RESPONSE: As already mentioned in one of the earlier points, we are developing implied emission factors for a number of sectors where changes in structure of the sector or increased penetration of control technology made an impact. Examples include heavy duty trucks where we estimate for BC implied emissions factor declined globally by nearly 20% (2010 to 1990) but in several regions like North America, Western Europe, Japan the reduction was over about 60-65%, Central Europe about 40-50%, but for Russia less than 10% and for most other regions no significant change was estimated (for several regions like China the impact of legislation is visible only in 2015 and later). Another example includes coal power plants where globally emission factors for PM2.5 dropped by about 45% with NA, Western

Europe, Japan having over 80% decline and even for China we estimated over 70% reduction while in Russia and several FSU countries only 20-30% decline. For industrial coal use lower reductions were achieved with exception of Eastern Europe and some FSU where collapse of heavy industry in the period 1990-2000 resulted in decline of emission factors by over 90% compared to the 1990 period. Finally, for residential heating (fuelwood) the 'global average factor' declined by about 15% which is mostly due to moderate changes in North America, Japan but mostly Western Europe where nearly 40% decline was estimated owing to strong increase in biomass use but in new installations including hundred thousands of pellet stoves and boilers. We will include brief discussion of these trends and illustrate it on a chart in the manuscript – see sections 221, 222,223,224 and figures 1,2,3,

REVIEWER:
Pg 14 - "the independent fuel estimate by Denier van der Gon et al. (2015).". van der Gon et al. state "A consistent set of PM10, PM2.5 and PM1 emission data for Europe was obtained from the GAINS model...", so these do not appear to be independent

RESPONSE: We need to point out that our statement refers to the 'independent fuel use' estimates by van der Gon et al. Indeed, they relied on GAINS emission factors but estimated fuelwood consumption independently rather than relying on statistical data. Therefore, we believe that this is an accurate statement.

REVIEWER:
pg 16 - "The resulting fuel use was compared and calibrated to the diesel consumption reported in the power and commercial sector." Not clear what this means.

RESPONSE: For DG sets, fuel use is estimated from the number and size of DG sets in some regions (i. e. Nepal, Nigeria). For some regions, share of diesel used in DG sets as a percentage of total diesel consumption in the country/region is available (i. e. India). Once the total diesel consumption by country/region is estimated then the fuel use for DG sets in GAINS model is allocated from commercial and power sector. We have revised the statement in the manuscript that reads now: "*The resulting fuel use was compared to the IEA statistics for the power and commercial sector and adjusted if necessary so that the overall energy use is consistent with the IEA.*"

REVIEWER:
pg 17, last portion of diesel generator section. There is some discussion of emission reduction options, but no mention the extent to which these were assumed to be applied in the emission estimates.

RESPONSE: Currently we have not applied any of the post-combustion control measures for DG sets in the developing world where they matter most; at least regionally. We modify that section and add explicit statement that no post-combustion measures are implemented for developing countries in this period.

The last paragraph of section 3.3 includes now the following sentence: "*While it is possible to achieve emissions reductions from diesel combustion through engine modifications and post-combustion measures, we assume that in the period 1990-2010 DG sets operating in the developing world lack any such controls. In case new information will become available and for future implementation of respective policies, the GAINS model includes a number of post-combustion control technologies such as diesel particulate filters (DPFs), diesel oxidation catalysts (DOC), and fuel-borne catalysts (FBC) offering reduction of gaseous and particulate emissions (Herzog, 2002; Yelverton et al., 2016)*."

REVIEWER:
pg 17 - line 25. IEA energy statistics contain separate lines for agriculture and construction - while these are not available for all years, these data seem to be becoming more complete in more recent years, even for non-OECD countries. It appears that this data is not used? Some further explanation would be useful.

RESPONSE: In fact we do make use of this information but this (IEA statistics) data is for total diesel consumption in agriculture or industry and not specifically in mobile machinery. For example, in agriculture diesel is used also for irrigation purposes (we attempt to allocate this to diesel generation sets in GAINS as the operating conditions are similar) but also in heating boilers used for example for drying or heating. The text in the manuscript does specifically refer to mobile machinery information and therefore we believe it is a valid statement.

REVIEWER:
pg 17 - line 31 "Also old and often poorly maintained vehicle fleet is reflected in measurements of emission factors" - not clear what is meant here.

RESPONSE: We agree, this statement is unclear and we replace it with: "*For all world regions we assume that a certain fraction of vehicles is badly maintained (e.g., Mancilla et al., 2012) or their emission controls tampered which is reflected as the share of so-called high-emitters (McClintock, 1999, 2007; Smit and Bluett, 2011; Yan et al., 2011, 2014); see further discussion in section 3.4.1.*"

REVIEWER:
pg 20 line 14. This "fuel consumption data for 2007 were extrapolated to 2010 using GDP". The result of this assumption should be compared to the fuel consumption estimates from IMO, who compared both bottom up and top down methodologies. ("Third IMO Greenhouse Gas Study 2014")

RESPONSE: We compared the result of our extrapolation for 2010 (1056 Mt $CO_2$ (13.83 EJ)) with the data in IMO 3rd GHG Study 2014 showing: [Table 1]

- For 2010: Total shipping: 915 mio t CO2 / International shipping: 771 mio t CO2
- Average 2007-2012: 1015 mio t CO2 / 846 mio t CO2

Our extrapolation is slightly higher than the average for the period 2007-2012 and about 10% higher than the reported fuel use in 2010 by IMO. We have added a remark and reference to IMO in the manuscript.

REVIEWER:
Page 23 top regarding coke ovens. It is not quite clear what the technology representation is here. "uncontrolled ovens" are mentioned, is the split between controlled and uncontrolled? Is this assumed to change over time? It appears there is little detail in terms of emission factors available in the literature (and emissions seem likely to depend on site specific characteristics in any event), so some comment might be useful on what are the most important data that would be needed to improve estimates.

RESPONSE: Indeed, coke industry PM emissions are poorly understood and this is of special importance for China in the last decade. We highlight the poor data availability with respect to measurements and add at the end a sentence about the assumption regarding the change in emission factor over time, i.e., "*Owing to lack of specific data for various world regions, we assume little change in emissions factors over time for the developing world, although the transition in China reported in Huo et al. (2012) was considered, and for OECD countries emission factor trend follows reported emissions, where available*."

REVIEWER:
Page 26. Might be useful to also mention that Agricultural waste burning can also be seasonally concentrated, so that it might be particularly important in come months.

RESPONSE: We have rewritten this part which now reads: "*At the same time, for several regions this source might be even more important, e.g., for Brazil we estimate its contribution at up to 15% of PM2.5 and 10% of BC emissions. Finally, agricultural burning has a strong seasonal pattern (see also section 2.4.1.) and has been also linked with heavy smog and haze episodes (e.g., Mukai et al., 2015; Stohl et al., 2007).*"

REVIEWER:
Page 26 line 12. "This database has been further extended and updated" perhaps edit to clarity that this refers to the data presented in the current paper.

RESPONSE: We have made that explicit now by joining this sentence and the next one that reads now: "*Niemi (2007) compared various datasets for all open biomass sources and developed the first global activity set for the RAINS model drawing on EDGAR3.2FT2000 (Van Aardenne et al., 2005) which we have* *further extended and updated to accommodate other data sources allowing gaps to be filled for several countries*."

REVIEWER:
Page 27 "3.9 Other sources" Is dust included? Its not mentioned until the discussion section

RESPONSE: We have added a sentence at the end of the first paragraph of this section: "*Note that windblown dust and emissions from unpaved roads are not included (see also introduction to section 3)*." As indicated here, there are few words of explanation about what is included and what not in the introductory section of the section 3 of the manuscript.

REVIEWER:
Page 28 line 13 - " for barbeques, a per capita emission factor is established, i.e.,...". Presumably this varies by region?

RESPONSE: Unfortunately, we found very little data allowing differentiating between the countries and therefore for most countries the same factor is used, except for few countries in Europe where national experts contributed their input leading to adjustment. We added a comment in the text reflecting that.

REVIEWER:
Page 32 line 19-20. Probably useful note here that the emissions in Granier et al., past about 2000/2005 were based on projections, where as the estimates here up to 2010 are (to the extent available) based on reported data and practices.

RESPONSE: Indeed, the numbers reported for 2010 represent various projections in Granier et al; in fact including also RCP numbers; this justifies also the widening emission range shown towards the end of the period they have investigated. We add a comment about it at the end of the line the reviewer referred to, which now reads: "….stabilization shown in earlier studies; *note that values reported in Granier et al. (2011) for 2010 were results of projections.*"

REVIEWER:
Page 35 line 3, this presumably is a typo? "confidence intervals to be 160-500% for the developing countries"

RESPONSE: As a matter of fact it is not a typo; see table 8 and discussion on page 18 of Streets et al (2003) manuscript. However, we choose to delete this statement from the paper as in view of the newer work (Bond et al, 2004, 2013), quoted already, where also a similar emission methodology was used as here, this particular statement does not add any specific insight unless elaborated further discussing specifically reasons for very high uncertainties estimated in that study.

REVIEWER:
Page 35 while "error compensation" is, indeed important, it might be useful to note that this might only be partial compensation. (e.g., some errors, such as measurement or enforcement issues, could be correlated across sectors.)

RESPONSE: Indeed, the poor enforcement might be one of the factors that cuts across the sector while for measurements of emission factors it could be also the case, often measurement techniques and teams performing them will be different reducing such potential. Our statement in the paper highlights the fact that the error compensation 'works' when errors are not correlated and so it is either known or assumed based on well-founded knowledge. We have added a respective comment in the paper and now the whole statement reads: "*Additionally, the error compensation, which is especially relevant if calculated emissions are the sum of a large number of equally important source categories (and where the errors in input parameters are not correlated with each other), can lead to a further reduction of overall emission uncertainty (Schöpp et al., 2005). A careful assessment of the assumption about correlation between input parameters is essential as for example poor enforcement of legislation or measurements errors could affect several source sectors in a similar way*."

REVIEWER:
Wasn't sure about the meaning of this sentence: "In fact, they could be even lower considering that they typically rely on a harmonized data set and include a simultaneous calculation of emissions of several species using the same principal activity and technology data." I would presume most country-level inventories are similar in this respect?

RESPONSE: Indeed, this statement is more relevant for the multipollutant inventory (whole GAINS framework) rather than PM alone. We delete this sentence in the final manuscript even though from our experience working with several regional and national inventories we see that often the methods applied to for example PM10 or PM2.5 are not the same as for BC or OC. The latter are often derived using simply shares of PM2.5 rather than absolute emissions factors representative for a given technology. Such shares are often derived from a large set of measurements representing a category of installations rather than a specific one for which PM2.5 emissions were calculated. In that way additional uncertainty is introduced.

REVIEWER:

Would be useful to mention in this section that there is also uncertainty in the speciation fractions (but this is constrained across species since these must sum to <= 1).

RESPONSE: Thank you for pointing it out, we add a sentence highlighting this point on page 35, line 9 [in the original submission] and it reads: "*Allocating total PM emissions into different size bins or chemical species (here BC and OC) is associated with uncertainties that for a specific source are determined by the measurement. Among others, Bond et al. (2013) discussed specific issues related to BC and OC aerosols, while for PM size distribution there exist specific analysis for particular measurement equipment (e.g., Armas et al.,2007; Coquelin et al., 2013) and most of the studies reporting measurements of size distribution estimate uncertainties for each size category. While the sum of all the PM species is constrained by the total mass, the single size distribution values rely on a large number of measurements reducing the overall uncertainty. Exceptions are source-sectors for which very few measurements exist, e.g., coke ovens, fireworks, handling of bulk materials.*"

REVIEWER:
Page 36 In addition to this "Our new global estimate of BC emissions suggests higher numbers than previously published...", perhaps useful to also mention something about BC trends over time here (since there is substantial interest in BC, and it looks like BC trends can be different from PM trends).

RESPONSE: More detailed discussion of this is actually provided in section 4.1 on page 30 of the original manuscript. Here, in conclusions we have added a statement about the different PM2.5 and PM10 trends vs BC and slightly reformulated the concerned paragraph (originally p.36, from line 10) and it reads now: "*We estimate that global emissions of PM have not changed much between 1990 and 2010 but there are significantly different regional trends with North America, Pacific, and Europe reducing emissions by 30 to over 50% and Asia and Africa increasing by about 30%. While these regionally varying developments are clearly visible in PM2.5 and PM10 estimates, the BC regional changes were somewhat less dramatic, mostly because trends in power and industrial sector emissions of PM are much less relevant for total black carbon emissions. Globally, over 75% of anthropogenic PM10 and PM2.5 originates from residential combustion, power plants and industry while for BC residential combustion and transport represent more than 75% but the importance varies across regions with Europe and North America having transport as key and rest of the world residential combustion. Our new global estimate of BC emissions suggests higher numbers than previously published owing primarily to inclusion of new sources*."

REVIEWER:
Supplement When GAINS values are listed in the tables, these are sometimes listed as ranges. I assume these are not uncertainty ranges (as in some of the other ranges in the table), but are GAINS central values and that the range represents the range used in different GAINS regions? It would be useful to clarify this.

RESPONSE: Indeed, these are ranges representing the spread of values across different regions rather than uncertainty ranges. While this is written for example on page 4 before the Table S2.1, we add a respective comment next to other tables in the SI.

REVIEWER:
I suggest providing a more detailed summary of the emissions data. It would make this data more readily useful to the community to have an electronic file (either csv or excel) that provides emissions of the various species by country/region and by sector and fuel (I appreciate that some aggregation with regard to sector/fuel might be necessary). I realize much of this (or perhaps all of this) would be available on-line, but providing this in a supplement will be more accessible and also provide for an archival record of these important results.

RESPONSE: As indicated in the responses to the initial comments of the reviewer, we have developed an additional set of tables with sectoral emissions (including split across key fuels) for the 25 global regions and all considered PM species over time. This is now included as the MS Excel file in the SI.

**RESPONSES TO Reviewer 2:**

We thank the reviewer for useful comments which have been helpful in improving the manuscript. The responses to specific points raised by the reviewer are provided below.

REVIEWER:
The title mentions that the paper focuses on anthropogenic emissions. However, the paper also discusses open fires. Since the paper is already very long, it might be better to only focus on anthropogenic emissions as stated in the title. The inclusion of emissions from fires (which come from other authors) is a bit confusing.

RESPONSE: Indeed, the paper documents the methodology for PM estimation in GAINS focusing on anthropogenic sources, including also open burning of agricultural waste, but at the same time documents also the complete dataset of ECLIPSE emissions. The latter includes explicit information about what has been used in modelling exercises (see for example Stohl et al., 2015; Eckhardt et al., 2016) and here the reference to the open fires, or more specifically forest and savannah fires is referred to. We allocate one section in the paper to agricultural burning (not all open fires) [see section 3.7] and make explicit references to work on forest and savannah fires on page 13 in the introduction to section 3. We feel that it is justified to provide full documentation of sources used in the entire ECLIPSE set, including forest savannah fires and allocate a page to discuss specific aspect of agricultural burning for which national and regional inputs were used beyond remote sensing data of GFED.

REVIEWER:
Abstract and line 24, page 2: the abstract claims this paper is " the first comprehensive assessment of historical (1990-2010) global anthropogenic particulate matter (PM): : :". However, the EDGAR4.3 inventory described in Crippa et al. (2016) provides emissions for 1970-2010 for PM2.5 and PM10. The statement about being "the first comprehensive assessment" is true for PM1, but not for the other species. Please rephrase.

RESPONSE: As a matter of fact, the word 'comprehensive' is referring here to the comprehensive assessment of several PM species (as well as forming the base for the development of the particulate number inventory referred to in the abstract) within one system assuring that consistent framework is used for the assessment of all of the considered species including PM1, PM2.5, PM10, PMTSP, BC, OC, and OM. But in order to avoid any possible misinterpretation or confusion we simply delete the word 'first' in the abstract as well as in the introduction.

REVIEWER:
Page 18, line 6: " exceptions are old vehicles running on leaded gasoline and preregulation 2-stroke mopeds : : : while latest gasoline direct injection engines have PM mass emissions comparable or even higher than latest diesel engines with particle

filter, however, the absolute level is about one order of magnitude lower than for older generations. This sentence is not clear. What does "absolute level" refer to?

RESPONSE: Thank you for pointing this out. We will rephrase this sentence making clear that the ‘absolute level’ refers here to the modern (Euro5 and Euro 6) diesel vehicles which have reduced their PM emissions significantly compared the pre or early control stages. Below an example for Italy to illustrate the point with COPERT data for emission factor for PM10 [in g/km]

| g/km | No-control | Advanced controls |
|---|---|---|
| Two-stroke | 0.176 | 0.018 (Euro 3) |
| Light duty gasoline | 0.0024 | 0.0010 (Euro 3 and younger) |
| Light duty diesel | 0.216 | 0.0018 (Euro 5 and younger) |

Interpretation:
=> old 2-strokes are as bad polluters as uncontrolled diesel cars.
Modern diesel cars have reduced their emission rate by a factor 100, such that they are today at the level of or even lower than modern gasoline cars.

The updated manuscript includes a modified sentence reflecting the above explanation. The new sentence in section 3.4 (middle of the 4th paragraph) reads: “*It is important to highlight that properly functioning particulate filters reduce PM emissions significantly and consequently the absolute level of the latest diesel vehicles is about two orders of magnitude lower than for older generations*.”

REVIEWER:
Page 26, line 20: The authors use quite old data for emission factors for agriculture waste burning. Akagi et al. (Atmos. Chem. Phys., 11, 4039–4072, 2011) have published a more recent and detailed review of all data available on emission factors. The authors should indicate why they did not use this more recent review.

RESPONSE: The paper by Akagi et al (2011) is included in the references and in fact we have considered it while comparing and deriving emission factors for this work. Our emission factors derived from several studies listed compare well with the ones provided in the review by Akagi since they mostly refer to the same work already listed in our previous text. We add explicit reference to Akagi paper in the revised text.

REVIEWER:
Page 29, lines 1-4: these lines should be rephrased. Many recent global chemistrytransport and chemistry-climate models now include detailed aerosols schemes, and PMs distribution are calculated as the sum of the mass of all the components included in the models. Maybe a few older models use the "BC + 1.4 OC" formula to calculate the mass of PM, but the recent models are much more advanced and calculate the mass of PMs in a more accurate way.

RESPONSE: Thank you for pointing this out, in fact we would be interested to know which models are these so that we could an example reference. The mentioned paragraph was meant to highlight two elements, the issue of oversimplifying the total carbonaceous mass in PM where often 1.4 ratio was used to convert OC to POM and the fact that the BC+POM often represents total fine anthropogenic PM in global (not regional) models. Taking into account your comments we have revised one of the sentences in this paragraph that reads now: "*This total fine PM mass has been typically estimated as BC+1.4*OC and only recently a number of models included more detailed aerosol schemes accounting for varying BC/OC ratios while still largely neglecting the anthropogenic dust component (e.g., Philip et al., 2017)*"

REVIEWER:
Page 31, lines 14-15: The sentence starting with "combined : : :" is unclear

RESPONSE: We have rewritten that sentence, specifically the second part starting with 'combined', and the whole sentence reads now: "*However, as further discussion shows, the largest discrepancy for $PM_{10}$ and $PM_{2.5}$ is for China as well as Europe and Russia; the sum of the differences in these three regions represents about 90% and over 50% of all the difference for $PM_{10}$ and $PM_{2.5}$.*"

REVIEWER:
Page 25 of the supplement: the authors should add in their table the TNO-MACC and TNO-MACCII (Kuenen et al., ACP, 2014) inventories, which provide emissions of PM for Europe and neighboring countries. The TNO-MACC inventories are now becoming a reference for atmospheric modeling in Europe, and these emissions should be mentioned in the paper.

RESPONSE: Thank you for this suggestion. We have included MACCII reference and also added the respective emission estimates to the table

REVIEWER:
Page 25 of the supplement: The emissions provided by US EPA are given as the sum of anthropogenic and wildfires. The dataset provided by EPA (note that the las release of the emissions is 2016 and not 2011 as mentioned in the supplement) provides emissions with and without wildfires. It would be better to include the emissions without wildfires, in order to be consistent with the other data in the table.

RESPONSE: Thank you for suggesting the review of this numbers. We retrieved new numbers from the US EPA website (https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei) for both 2011 and 2014. This allows now for a better comparison to our numbers constructing a similar sector set excluding wildfires. The reference to this EPA source is also included in the manuscript.

# Global anthropogenic emissions of particulate matter including black carbon

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**Abstract.** This paper presents a comprehensive assessment of historical (1990-2010) global anthropogenic particulate matter (PM) emissions including the consistent and harmonized calculation of mass-based size distribution ($PM_1$, $PM_{2.5}$, $PM_{10}$), as well as primary carbonaceous aerosols including black carbon (BC) and organic carbon (OC). The estimates were developed with the integrated assessment model GAINS, where source- and region-specific technology characteristics are explicitly included. This assessment includes a number of previously unaccounted or often misallocated emission sources, i.e., kerosene lamps, gas flaring, diesel generators, trash burning; some of them were reported in the past for selected regions or in the context of a particular pollutant or sector but not included as part of a total estimate. Spatially, emissions were calculated for 170 source regions (as well as international shipping), presented for 25 global regions, and allocated to 0.5° x 0.5° longitude-latitude grids. No independent estimates of emissions from forest fires and savannah burning are provided and neither windblown dust nor unpaved roads emissions are included.

We estimate that global emissions of PM have not changed significantly between 1990 and 2010, showing a strong decoupling from the global increase in energy consumption and, consequently, $CO_2$ emissions but there are significantly different regional trends, with a particularly strong increase in East Asia and Africa and a strong decline in Europe, North America and the Pacific region. This in turn resulted in important changes in the spatial pattern of PM burden, e.g., European, North American, and Pacific contributions to global emissions dropped from nearly 30% in 1990 to well below 15% in 2010, while Asia's contribution grew from just over 50% to nearly 2/3 of the global total in 2010. For all ~~considered~~ PM species considered, Asian sources represented over 60% of the global anthropogenic total, and residential combustion was the most important sector, contributing about 60% for BC and OC, 45% for $PM_{2.5}$ and less than 40% for $PM_{10}$ where large combustion sources and industrial processes are equally important. Global anthropogenic emissions of BC were estimated at about 6.6 and 7.2 Tg in 2000 and 2010, respectively, and represent about 15% of $PM_{2.5}$ but for some sources reach nearly 50%, i.e., for the transport sector. Our global BC numbers are higher than previously published owing primarily to the inclusion of new sources.

This PM estimate fills the gap in emission data and emission source characterization required in air quality and climate modelling studies and health impact assessments at a regional and global level, as it includes both carbonaceous and non-

carbonaceous constituents of primary particulate matter emissions. The developed emission data set has been used in several regional and global atmospheric transport and climate model simulations within the ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) project and beyond, serves better parameterization of the global integrated assessment models with respect to representation of black carbon and organic carbon emissions, and built a basis for recently published global particulate number estimates.

## 1 Introduction

Particulate matter (PM) or aerosols are solid and liquid particles small enough to remain airborne. PM can be directly emitted to the atmosphere (primary PM) or it can form from gaseous precursors (secondary PM). The size of PM stretches from clusters of molecules with a diameter of a few nanometers up to micrometer-sized abrasion products. This vast dimensional spectrum is reflected in the varying composition and characteristics of PM measured at source and receptor sites. PM species are important constituents of the atmosphere and they play a role in the earth's climate system. Some PM species, i.e. black carbon, absorb visible light and warm the atmosphere, whereas other species, i.e., sulphates and organics reflect sunlight back to space and cool the climate (Bond et al., 2013). PM also serves as condensation nuclei for water vapour to eventually form cloud droplets. There is well-documented evidence that exposure to PM results in adverse effects on human health (e.g., Anenberg et al., 2012; Lim et al., 2012; WHO, 2004).

Integrated assessment models, such as the GAINS (Greenhouse gas - Air pollution Interactions and Synergies) model (Amann et al., 2011), utilize data on economic development and corresponding pollutant emissions, estimate atmospheric concentrations and further assess the impacts on climate, human health and ecosystems. When this information is combined with potentials and costs for controlling the emissions, it is possible to study the cost-efficiency of different policies to reduce the undesirable effects and meet environmental objectives on climate, human health and ecosystem impacts. Such an integrated modelling framework is particularly important for assessing the impacts of particulate matter owing to the multitude of sources, including primary and secondary, and effects on health and climate. All these aspects of PM call for consistent data to support the assessments of impacts and potential for formulating robust strategies to reduce emissions together with consequent concentrations and impacts.

This paper presents a comprehensive assessment of historical (1990-2010) global anthropogenic particulate matter (PM) emissions including the consistent and harmonized calculation of mass-based size distribution ($PM_1$, $PM_{2.5}$, $PM_{10}$), as well as primary carbonaceous aerosols~~; –~~, black carbon (BC) and organic carbon (OC). The methodology draws on the earlier developed structure of the PM module in GAINS (Klimont et al., 2002b; Kupiainen and Klimont, 2004, 2007) but was extended to include new information as well as sources previously unaccounted ~~sources~~for, i.e., gas flaring, kerosene lamps, and diesel generators.

A recent GAINS model development ~~includes extension~~extends its scope to include particulate number (PN) emissions (Paasonen et al., 2013). This builds on the emission methodology and estimates described in this paper, making use of one of the datasets (ECLIPSE *V5*) to calculate past and future PN emissions and their spatial distribution. The respective documentation and discussion paper is available in ~~review~~ (Paasonen et al., 2016).

While the results presented in this paper focus on the outcomes included in the ECLIPSE *V5a* version of the data, there were several datasets developed within the ECLIPSE project[1] (Stohl et al., 2015) and the key differences between the datasets are also briefly discussed. Table 1 gives an overview of the datasets that are accessible from the GAINS website[2]; the paper describing the projections is in review for this issue of ACP (Klimont et al., in preparation).

## 2 Method

The ECLIPSE emission data set was created with the GAINS (Greenhouse gas – Air pollution Interactions and Synergies; http://gains.iiasa.ac.at) model (Amann et al., 2011), which calculates emissions of air pollutants and Kyoto greenhouse gases (GHG; i.e., carbon dioxide ($CO_2$,), methane ($CH_4$), nitrous oxide ($N_2O$) and the three F-gases) in a consistent framework. The GAINS model holds essential information about key sources of emissions, environmental policies, and further mitigation opportunities for 170 country-regions. The model relies on international and national statistics of activity data for energy use, industrial production, and agricultural activities (see section 3) for which it distinguishes all key emission sources and control measures. Several hundred technologies to control air pollutant and greenhouse gases emissions are represented, allowing simulation of implemented air quality legislation (see section 2.3).

Since previous work (Cofala et al., 2007; Klimont et al., 2002b, 2009, Kupiainen and Klimont, 2004, 2007; Shindell et al., 2012) we have reviewed recent literature, including non-peer reviewed studies, to improve characterization of the source sectors and control technologies in the GAINS model, update the assumptions about penetration of control measures, and to include previously unaccounted or poorly allocated sources. Emission sources that have been recently added, or for which the emission calculation has been refined, include~~:~~ flaring of associated petroleum gas in the oil and gas exploration sectors, kerosene lamps for lighting (further development of estimates originally presented by Lam et al. (2012)), diesel generator sets, ~~high~~ high-emitting vehicles, international shipping, trash burning ~~as well as~~and brick kilns (see section 3).

Further improvements in the emission model have been made especially for China (Klimont et al., 2013; Wang et al., 2014; Zhao et al., 2013), where large changes have occurred recently as well as new data becoming available, but also for Europe where results of the consultation with national experts during the review of the EU National Emission Ceilings Directive were considered in the last datasets (Amann et al., 2015). Finally, the regional resolution of the global GAINS model has been improved by distinguishing more countries in Latin America where five regions (Argentina, Brazil, Chile, Mexico, all remaining Latin America) were replaced with 13 regions in version *V5a*, including most countries of South America,

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[1] European Commission FP7 project ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants); Project no. 282688; http://eclipse.nilu.no

[2] http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html

Mexico, Central America, and the Caribbean; a full list of country-regions in the global GAINS application is included in the supporting information (SI).

### 2.1 PM estimation method

The methodology to derive particulate matter (PM) emission factors and calculate emissions relies on the methods documented in (Klimont et al., 2002b; Kupiainen and Klimont, 2004, 2007). However, apart from updates to emission factors a number of modifications and extensions have been introduced ~~since~~subsequently, especially for carbonaceous particles. We summarize the principles below, allocating more space to discuss extensions.

The emissions of PM in the GAINS model are calculated for several size classes: a submicron fraction (particles with diameter smaller than 1 µm; $\leq PM_1$), a fine fraction ($\leq PM_{2.5}$), a coarse fraction ($>PM_{2.5}$, $<$~~>~~$PM_{10}$), and large particles ($\geq PM_{10}$). $PM_{10}$ is calculated as the sum of fine and coarse fractions~~.~~, ~~Total~~ total suspended particles (TSP) as the sum of fine, coarse, and $\geq PM_{10}$ fractions. Additionally, black carbon (BC) and organic carbon (OC) are calculated.

The methodology includes the following steps:

(i) region- (*i*), sector- (*j*) and fuel- (*k*) specific "raw gas = unabated" emission factors for total suspended particles (TSP) are derived. For solid fuels (excluding biomass and use of solid fuels in small residential installations) the mass balance approach is used where ash content (*ac*) and heat value (*hv*) of fuels, and ash retention in boilers (*ar*) for given combustion technologies are considered Eq. (1):

$$ef(TSP)_{i,j,k} = \frac{ac_{i,j,k}}{hv_{i,j,k}}(1 - ar_{j,k}) \tag{1}$$

For liquid fuels, biomass, solid fuels used in small residential installations, industrial processes, mining, storage and handling of bulk materials, waste incineration, agriculture[3], and transport, TSP emission factors are taken from the literature;

(ii) considering fuel- and sector-specific size fraction profiles reported in the literature, "raw gas" emission factors for each of the size fractions and carbonaceous species are estimated;

(iii) the emission factors for organic carbon (OC), calculated in the previous step, are adjusted considering the carbonaceous fraction in $PM_{2.5}$ and organic carbon (OM); see section 2.1.1 for discussion;

(iv) PM emissions are calculated for each size fraction and carbonaceous species applying the following equation Eq. (2), where also the application rates of control technologies (*X*) and size fraction specific emission removal efficiencies (*eff*) are taken into account:

$$E_{i,y} = \sum_{j,k,m} E_{i,j,k,m,y} = \sum_{j,k,m} A_{i,j,k}\, ef_{i,j,k,y}\,(1 - eff_{m,y})\, X_{i,j,k,m}\,, \tag{2}$$

where *i*,*j*,*k*,*m* are region, sector, fuel, abatement technology; *y* size fraction, i.e., fine, coarse, PM_>10, or carbonaceous species (BC, OC); $E_{i,y}$ emissions in region *i* for size fraction *y*; *A* activity in a given sector, e.g., coal consumption in power

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[3] For livestock, emission factors refer to housing period and, therefore, information on the length of this period (one of the parameters in the GAINS model) is considered to derive annual animal- and country-specific values.

plants; *ef* "raw gas" emission factor; $eff_{m,y}$ reduction efficiency of the abatement option *m* for size fraction *y*, and *X* actual implementation rate of the considered abatement, e.g., percent of total coal used in power plants that are equipped with electrostatic precipitators. If no emission controls are applied, the abatement efficiency equals zero ($eff_{m,y} = 0$) and the application rate is one ($X = 1$). In that case, the emission calculation is reduced to simple multiplication of activity rate by the "raw gas" emission factor.

There are a few source sectors where additional assumptions are made in order to develop emission factors used in the calculation. Specifically, for gas flaring additional information about the composition of associated gas is used (see section 3.6.3 for more details), and to estimate emissions from ~~high~~ high-emitting vehicles (or super-emitters) assumptions about region-specific shares of high emitters as well as technology and ~~pollutant~~ pollutant-specific increments, compared to the average fleet emissions factors (excluding high emitters), are made (see section 3.4.1).

### 2.1.1 Adjustments of carbonaceous particle emission factors

While we principally follow the definition of black carbon (BC) given by Bond et al. (2013), i.e., *"…a distinct type of carbonaceous material that is formed primarily in flames, is directly emitted to the atmosphere, and has a unique combination of physical properties. It strongly absorbs visible light, is refractory with a vaporization temperature near 4000K, exists as an aggregate of small spheres, and is insoluble in water and common organic solvents"*, the available measurement studies have not been consistent in this respect, and it has not been possible to systematically follow the definition in developing the input data for emission estimates; this has also been discussed in our previous papers (Kupiainen and Klimont, 2004, 2007).

Organic carbon (OC) refers to the carbon fraction in numerous organic compounds that contain hydrogen and, usually, oxygen, and are emitted to the air as particles (Bond et al., 2013). To attain the total mass associated with the organic compounds, organic matter (OM), OC needs to be multiplied ~~with~~ by a fraction that depends on the suite of compounds emitted and varies between ~~the~~ emission sources. We introduce ~~source~~ source-specific OM to OC fractions for primary emissions found from literature, varying between 1.3 and 2.1 (Aiken et al., 2008; Tissari et al., 2007; Turpin and Lim, 2001). Due to the lack of a formal definition and available measurement studies we have not attempted so far to separate emissions of "brown carbon", a group of absorbing compounds considered a subset of organic aerosol (Bond et al., 2013).

Emission factors of organic carbon ($ef_{OC}$) for each GAINS technology category are calculated using a mass balance equation Eq. (3). This equation has been introduced to ensure that the mass balance of the chemical species of particulate matter (black carbon and organic carbon) will still stay within physical limits of the PM mass metrics applied in GAINS. The calculation uses $PM_{2.5}$ as the limiting mass metric since the emissions of carbonaceous matter occur primarily in that size range. We introduce only a few exceptions where larger carbonaceous particles are expected to be present, e.g., tyre wear.

$$ef_{OC} = (ef_{PM2.5} \times f_{carb} - ef_{BC}) \div f_{OM}, \qquad (3)$$

where $f_{carb}$ is the mass fraction of the total carbonaceous matter, or black carbon and organic matter, in $PM_{2.5}$, $f_{OM}$ the average organic molecular weight per carbon weight in particular matter, $ef_{BC}$ the emission factor of BC, $ef_{PM2.5}$ the emission factor of

$PM_{2.5}$. Emission factors of BC and $PM_{2.5}$ as well as $f_{carb}$ and $f_{OM}$ are estimated based on emission measurement data. The final set of OC emission factors is checked for consistency with emission measurements.

The f~~F~~raction of carbonaceous matter in $PM_{2.5}$ ($f_{carb}$) varies significantly between source sectors. Highest fractions are usually found in residential combustion and transport sectors in technologies with poor combustion, where over 90 percent of the particulate matter is estimated to consist of carbonaceous matter. As the combustion process becomes more efficient and optimized, the fraction reduces drastically and, for example, in large modern power plants, which have optimized combustion processes and efficient air pollution abatement technologies, the fraction is typically negligible; see discussion in Kupiainen and Klimont (2007) and Sippula et al. (2009).

The average fraction of organic molecular weight per carbon weight ($f_{OM}$) also varies ~~also~~ between different emission source sectors and fuels. For combustion of biomass, including wood, we use $f_{OM} = 1.8$, which represents approximately the middle of the range (1.6 to 2.1) of $f_{OM}$ values available for combustion of different wood species in the literature (Aiken et al., 2008; Tissari et al., 2007; Turpin and Lim, 2001). For diesel and gasoline in transport sector, we use $f_{OM} = 1.3$, based on Aiken et al. (2008).

### 2.2 Model technology resolution

The GAINS model structure includes representation of key emission sources compatible with global and regional emission inventories but the calculation often distinguishes an additional level of detail where combustion technology (e.g., pulverized coal or grate firing boilers, fireplaces, various stoves, pellet boilers, etc.) as well as emission control technology (e.g., wet scrubbers, fabric filters, fan assisted stoves, diesel particulate filters, etc.) are explicitly distinguished (see also Eq. (2)). Such an approach has been an integral part of the GAINS model development for both particulate matter (e.g., Klimont et al., 2002b; Lükewille et al., 2001) and other pollutants (e.g., Amann et al., 2011; Cofala and Syri, 1998; Klimont et al., 2002a); the details for PM are documented in Klimont et al. (2002b) and the current structure can be reviewed in the on-line application of the GAINS model[4]. This approach has also been used in other emission assessment studies and is often referred to as 'technology-based' (e.g., Bond et al., 2004; Lu et al., 2011; Zhao et al., 2013).

Implementation of such technology resolution requires additional assumptions about the shares of activity in a given sector falling into each subcategory and the share of activity controlled with a specific mitigation measure. The following sections highlight and document briefly the assumptions for key sectors.

#### 2.2.1 Residential combustion: cooking, heating, lighting

GAINS divides the residential-commercial sector into several fuel–dependent categories (Table 2). The division is driven by varying emission characteristics and available control options (Table 3). While such a structure is fairly compatible with the available emission measurements (see section 3.1), it is challenging to distribute fuel consumption into these categories as

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[4] http://gains.iiasa.ac.at; select any of the accessible regional versions to view the model structure

typically statistical data ~~is~~ are available either as total residential sector or split into commercial/residential/other (e.g., IEA, 2015a, 2015b). We rely on a mix of sources and our own assessment to derive the respective shares of technologies, which change over time. There have been several assessments at a global level where either total fuel demand for cooking and heating, allocation between various fuels, or stove types was attempted (Bonjour et al., 2013; Chafe et al., 2014; Fernandes et al., 2007). For Europe, such data are not readily available; however, within the work on the revision of air quality legislation we were involved in several rounds of stakeholder consultations where national experts representing various sectors reviewed GAINS assumptions (Amann et al., 2015) and all data can be viewed in the on-line model. Additionally, information about pellets and pellet stoves and boiler sales (e.g., Paniz and Bau, 2014; WIP, 2009) resulted in adjustment of shares of biomass used in such installations in several European countries where strong growth has been observed towards the end of the period under investigation. For the US and Canada, a similar discussion and exchange took place within the work of the Arctic Council where the GAINS model was used to develop unified emissions and scenarios (AMAP, 2015). For Australia and New Zealand a number of local studies were used (Driscoll et al., 2000; Scott, 2005; Todd, 2003). Also for China, trends towards cleaner coal stoves (e.g., Zhi et al., 2009) and more household coal boilers (in specific provinces) were taken into account.

The allocation of fuel between various categories varies between Europe, North America, and OECD Asia and Pacific where most solid fuel is used for heating (e.g., Chafe et al., 2015), and most of Asia, Africa and Latin America where cooking is the primary use. Consequently, nearly all solid fuels in South Asia, Africa~~,~~ and Latin America are allocated to cooking stoves. For Asia, we draw on the past and ongoing collaboration on the development of the GAINS-Asia model (Amann et al., 2008; Klimont et al., 2009; Zhang et al., 2006; Zhao et al., 2013) where assumptions on the split between heating and cooking, as well as fuel used in medium size boilers were made, ~~as well as~~together with several peer-reviewed publications (e.g., Aggarwal and Chandel, 2004; Venkataraman et al., 2010)~~(e.g., Venkataraman et al., 2010)~~. For Latin America, information about this sector structure originates from ~~the~~ discussions with the authors of various assessments of effectiveness of clean cooking programs (e.g., Pine et al., 2011; Ruiz-Mercado et al., 2011) as well as the data collected within the CCAC (Climate and Clean Air Coalition) and UNEP supported Integrated Assessment of Short-Lived Climate Pollutants in Latin America and Caribbean[5] (final report is in preparation for publication; see summary for policy makers). The ratio of cooking/heating is assumed constant in the 1990-2010 period as we have not found any data allowing to change that assumption.

The GAINS model includes a number of mitigation measures in this sector (Table 3), although some of them might be seen more as different types of installations, e.g., various stove types already in place (for specific discussion of their assumed characteristics see Supplementary Information – section S2). While there has not been a lot of success in sustained replacement of traditional stoves with improved clean burning stoves (e.g., Foell et al., 2011; Pine et al., 2011; Ruiz-Mercado et al., 2011; Wickramasinghe, 2011), it is important to consider the varying level of implementation across the regions if such information is available. As with the allocation of fuel use (see discussion above), we rely on data and

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[5] http://www.ccacoalition.org/en/resources/integrated-assessment-short-lived-climate-pollutants-latin-america-and-caribbean-summary ; publication of the final report is expected in 2017 and it will be available from the CCAC and UNEP website.

assessments collected within several bilateral projects (e.g., Amann et al., 2008, 2015), peer-reviewed papers (e.g., Klimont et al., 2009; Lewis and Pattanayak, 2012; Li et al., 2016; Pine et al., 2011; Ruiz-Mercado et al., 2011; Shrimali et al., 2011; Silk et al., 2012; Troncoso et al., 2011), and published reports (Adria and Bethge, 2013; Germain et al., 2008; Scott, 2005; Todd, 2003). Technology structure has an impact on the implied (average) emission factor for a given category distinguished in the model. While changes for biomass cooking stoves were rather limited at a larger scale, resulting in up to 10% decline in implied $PM_{2.5}$ emission factor in Asia and up to 5% in Latin America, we estimate a larger impact for residential biomass heating. We estimate that for $PM_{2.5}$, the 'global average emission factor' declined from 1990 to 2010 by about 15% which is mostly due to a strong increase in sales of pellet stoves and boilers in Western Europe leading to nearly 40% reduction in implied emission factor (Fig. 1). Interestingly, the changes in emission factors for BC are less pronounced (Fig. 1) since the improved stoves are more efficient in reducing the total level of particulate matter emissions rather than black carbon (see further discussion in section 3.1 and SI (S2).

One of the recent developments in the GAINS model was the explicit distinction of ~~use of~~ kerosene use between ~~for~~ cooking and lighting (Table 2); earlier all kerosene was allocated to cooking. This modification was driven by the study highlighting the potentially high contribution of kerosene lamps to black carbon emissions (Lam et al., 2012). The emissions depend on what type of lamp is used, and for historical data we distinguish between wick and hurricane lamps, with the former representing the majority (Lam et al., 2012; Mills, 2005). As a default, we assume 80% kerosene wick lamps in South Asia and 50% in other developing world regions. For a discussion of how total activity data for kerosene lighting is calculated see section 3.2.

### 2.2.2 Transport

The GAINS model distinguishes several source categories within the road and non-road transport sectors. Road transport is disaggregated into six vehicle categories: 2-stroke/~~and~~ 4-stroke two-wheelers, passenger cars and vans, light duty vehicles, heavy duty trucks, and buses. The non-road mobile sources are grouped into ~~nine~~ eight broad categories: agriculture and forestry, construction and mining, rail, inland navigation, coastal shipping, aviation (only landing and take-off), 2-stroke engines (e.g., in households, recreation, forestry, etc.), and other ~~land~~ land-based engines. Each vehicle/machine category is associated with a fuel according to its propulsion type; several fuels are distinguished: diesel, gasoline, CNG, LPG, jet fuel or kerosene, heavy fuel oil, as well as hydrogen and electricity. For each of the fuel-vehicle combinations, activity data (fuel consumption and km-driven for road vehicles) are sought and are usually available in national and international statistics for road transport categories, while they are often incomplete, allocated under other sectors, or even lacking for non-road sources. For a complete list of transport sources and fuels see Table S8.1.

While we do not specifically model ~~specifically~~ vehicle vintages, the new emission standards are typically synonymous with a new vintage year of a particular vehicle category. In order to reflect existing legislation (section 2.3), each fuel-vehicle combination is further subdivided by its average emission level. The key proxy for the emission level is the exhaust emission legislation in force in the country (or region) at the time when the vehicle type is put into service or to which level it is retrofitted. The associated emission factors describe the emission rates for the pollutants averaged over the actual operating conditions, vehicle sizes, and machine types, as well as ages and model years within one emission standard. More details about the emission factors, control stages in GAINS, and discussion of ~~high~~ high-emitting vehicles are provided in section 3.4.

Depending on the region, the implied (average) emission factors for key vehicle categories have been changing ~~in~~ over the period considered ~~period~~. We estimate that by 2010 the global average BC emission rate has declined ~~by 2010~~ by nearly 20% for heavy~~ and ~~-duty vehicles but in several regions like North America, Western Europe, developed Asia and Pacific the reduction was about 60-65%, Central Europe about 40-50%, while for most other regions small or no significant change was estimated (Fig. 2). Similar trends were found for ~~light~~ light-duty vehicles but the reductions are typically higher with a global average declining by nearly 35% (Fig. 2).

### 2.2.3 Large scale industrial combustion

The available statistical data allow~~s~~ for allocation of fuel into key sectors, like power plants and industrial boilers, but owing to varying emission characteristics and often different legislation for different boiler types, the GAINS model distinguishes additionally a number of selected plant and boiler types (for more background discussion see Klimont et al. (2002b)). Specifically, the power sector is divided into existing (constructed before 2005), new and modern plants, for which additionally large and small plants (grate firing) are distinguished. Structural changes as well as increasing stringency of emission legislation resulted in declining emission factors. For example, we estimate that the global average PM$_{2.5}$ emission factor for coal power plants dropped by about 40%, with Northern America, Europe, and Japan having a 70-80% decline and even for China we estimate over 70% reduction, while in Russia and several Former Soviet Union countries only 20-30% decline (Fig. 3) is seen. Industrial combustion is associated with several sectors for which ~~also~~ small boilers are also included to capture the large numbers of often old and poorly controlled solid fuel ~~grate~~ grate-firing boilers in the developing countries ~~(e.g., Wang et al., 2014; Zhao et al., 2013),~~ (e.g., Wang et al., 2014; Zhao et al., 2013); for example, in China they accounted for about 85% of all industrial boilers (Wang et al., 2009). For industrial coal use lower reductions in average emission factors were achieved than for power plants, with the exception of Eastern Europe and some Former Soviet Union countries where the collapse of heavy industry in the period 1990-2000 resulted in a decline of emission factors by over 90% compared to ~~the~~ 1990. While the estimated changes in emission characteristics could be modelled more accurately if assumptions about equipment vintages were made, the GAINS model does not explicitly include ~~it~~ that information ~~with~~ except~~ion~~ for the ~~of~~ power sector (see above). Instead, GAINS defines technical lifetimes of the add-on control technologies (e.g., cyclones, electrostatic precipitators, fabric filters) and considers that these can be principally applied shortly after the respective legislation is put in place. Finally, the GAINS model structure has been extended to distinguish diesel generator sets; previous GAINS regional and global assessments of PM or carbonaceous particles (Cofala et al., 2007; Klimont et al., 2009; Kupiainen and Klimont, 2007) included their fuel consumption in the power and residential combustion sectors. The new structure allows for better representation of emissions and mitigation opportunities, especially in regions with low reliability of electricity supply and poor emission standards, e.g., South Asia. The estimates of regional diesel generators fuel use is discussed in section 3.3.

### 2.2.4 Industrial processes

Most industrial processes are sources of particulate matter emissions. For the majority of them emissions are calculated using total production volumes without distinguishing specific stages of the processing chain. However, for a number of manufacturing processes we define a default plant profile and distinguish between process and fugitive emissions, for details see Klimont et al. (2002). Additionally, for selected industries a more detailed structure was designed to reflect the significant differences between types of plants (kilns); this has been done for cement, coke, and brick manufacturing.

The key driver behind the extended structure for cement and coke manufacturing was developments in China where in the last decades strong growth resulted in often rapid transformation of the two sectors. For cement production rotary kilns with precalciner and shaft kilns are distinguished, for which the activity split has been developed in collaboration with Tsinghua University (Zhao et al., 2013). Such technological changes, often accelerated by political and economic transformation (e.g., Eastern Europe and Former Soviet Union), and the legislation landscape resulted in rather significant changes in average emission rates in the cement production sector. We illustrate that in Fig. 3 where in several regions GAINS implied $PM_{2.5}$ emission factors in 2010 are lower by up to 90% than in 1990. The coke production sector in China experiences rapid transformation from traditional ovens to mechanized integrated coke ovens which have different emission characteristics; the changes in the structure of the sector are discussed by Huo et al. (2012). Currently, the information about the comparable technology split is not available for other countries, for which emissions are calculated without such distinction.

*Brick manufacturing*

There are strong regional differences in the brick manufacturing sector structure that is especially relevant in the developing world where a large share of the market is occupied by traditional, heavy-polluting kilns. Our earlier work focused on characterizing the brick sector in Asia, by far the largest producer, and therefore the distinguished kiln types reflected practices in Asia (Klimont et al., 2009; UNEP/WMO, 2011). However, such a model design ~~of the model~~ did not allow to correctly address the structure of this sector in other regions like Africa or Latin America and the Caribbean. We have reviewed regional and national assessment studies to identify typical regional profiles (distribution of production by kiln types) of the brick manufacturing sector, including also typical~~ly used~~ fuels; such profiles change over time and ~~it~~ this has been considered where such information was found. Table 4 shows the kiln structure included in GAINS and highlights key representative technologies assumed for different world regions. The overview of studies used to develop the respective assumptions is provided in the SI (S5). The overall brick production data are discussed in section 3.6.2 and SI (Table S5.2).

## 2.3 Emission legislation

We have collected information about existing international and national requirements with respect to emission limit values for stationary and mobile sources and estimated control technology implementation rates required to achieve the respective standards in all GAINS regions. The interpretation of the laws and translation into the set of GAINS technologies with the associated emission rates under average operating conditions has been discussed previously in a number of papers and assessments addressing regional (Amann et al., 2015; Klimont et al., 2009; Kupiainen and Klimont, 2007; Wang et al., 2014) and global (Amann et al., 2013; Cofala et al., 2007; Rao et al., 2013; Riahi et al., 2012; UNEP/WMO, 2011) emissions.

For a number of sources there exist global databases summarizing current laws and emission limit values, including power plants (IEA, 1997; IEA CCC, 2012), transport (Delphi Inc., 2013, 2015; ICCT & Dieselnet, 2014), and the cement industry (Edwards, 2014). Additionally, specific regional and national laws and policy implementation studies were reviewed, i.e., for the European Union a number of Directives ~~was~~ were considered (Crippa et al., 2016; EC, 2001a, 2001b, 2010; Krasenbrink and Dobranskyte-Niskota, 2008), for Asia several peer-reviewed studies (Goel and Guttikunda, 2015; Guttikunda and Jawahar, 2014; Huo et al., 2011, 2012; Klimont et al., 2009; Liu et al., 2015; Lu et al., 2011; Wang et al., 2014; Zhang et al., 2006) as well as other sources (CAI-Asia, 2011; CPCB, 2007; IIDFC, 2009); for Latin America and Caribbean additional information was obtained for the brick sector (e.g., Stratus Consulting, 2014) and for Argentina, Brazil, and Mexico also for the transport sector (e.g., Ministério do Meio Ambiente, 2011).

In the course of development of the several ECLIPSE datasets, the legislation information and mostly the rates of enforcement and implementation of actual measures have been revisited. The key updates in version *V4a* (see Table 1) include consideration of the initial round of consultations with European Union member states' experts within the review of the National Emission Ceiling (NEC) directive (Amann et al., 2012), which included comparison of GAINS estimates with the emissions officially reported to the Centre on Emission Inventories and Projections (CEIP; www.ceip.at) under the Convention on Long-range Transboundary Air Pollution. A much more substantial update came with version *V5a* where for China the 12th Five Year Plan policies were introduced, resulting in revision of the implementation and enforcement rates of control measures for 2010, drawing also on analysis of progress in legislation implementation in China (e.g., Lin et al., 2010; Zhang et al., 2015). Furthermore, the legislation for the cement industry was reviewed and updated (Edwards, 2014), emissions from international shipping were also calculated, the treatment of ~~the~~ non-road mobile machines ~~were~~ was reviewed, and for Latin America and Caribbean (LAC) the GAINS model has been revised to include nearly all single countries[6] and, consequently, required definition of control strategies reflecting current legislation for each country. Finally, also for the European Union an update was performed in *V5a* to include the latest status of discussion with the national experts (Amann et al., 2015), as well as new submissions of $PM_{2.5}$ emissions (also for the past years) to CEIP, especially for 2010.

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[6] Previous versions included five regions: Argentina, Brazil, Chile, Mexico, other LAC

### 2.4 Spatial and temporal distribution

The GAINS model calculation is performed for 170 regions globally and for Europe and Asia the calculation and results are directly available by country or even subnational level from the online version of the model (http://magcat.iiasa.ac.at) for all ECLIPSE data sets. At a global level, the emissions and activity data are available online at the resolution of 25 global regions (see Supporting Information (SI), S7) and key sources (http://magcat.iiasa.ac.at/gains/IAM/index.login); the structure is compatible with most of the global integrated assessment models. Additionally, the total annual emissions were gridded and temporal (monthly) distributions were developed.

The GAINS particulate matter emissions were distributed into $0.5^{o}$ x $0.5^{o}$ longitude-latitude grids and stored in *netCDF* format files available from http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html as well as from the ECLIPSE project web: http://eclipse.nilu.no. The files contain several layers (Table 5), reflecting key sectors (consistent with Representative Concentration Pathways (RCP) used in the Intergovernmental Panel for Climate Change Fifth Assessment Report (IPCC AR5)), and a total emission layer. The spatial distribution was prepared from RCP-consistent proxies as used and further developed within the Global Energy Assessment project (GEA, 2012). These are in line with proxies applied within the RCP projections as described in Lamarque et al. (2010) and were modified to accommodate more recent information where available, e.g., population distribution, open biomass burning, effectively making them year-specific (Klimont et al., 2013; Riahi et al., 2012).

In the process of preparing gridded emissions we have developed additional layers which were merged into the sector layers listed in Table 5. The primary example, relevant for particulate matter emissions, is the flaring layer which has been developed by IIASA using the information on flare location areas developed in the collaborative project of NOAA, NASA, and the World Bank (Elvidge et al., 2009, 2011). This layer contains emissions from flaring in oil/gas exploration and it is for the first time that a global PM emission assessment includes this source with explicit spatial allocation (Fig. 4); this dataset was used within the ECLIPSE project and highlighted the relevance of proper distribution of black carbon emissions from this source (Stohl et al., 2013). The flaring emissions are integrated in the *Energy* (Table 5) layer but a separate file with all emissions from flaring only is also available for download.

#### 2.4.1 Temporal distribution

The GAINS model does not explicitly include any assumptions about temporal distribution and therefore all emissions are calculated as annual totals. However, within the MACEB[7] and ECLIPSE projects we have developed monthly emission profiles for the gridded output, shares of emissions in different months in each grid, for a number of sources. The focus was on allocation of domestic heating and cooking emissions where the methodology combines the stove use assumptions from

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[7] MACEB - **M**itigation of **A**rctic warming by **C**ontrolling **E**uropean **B**lack carbon emissions, European Union Life+ project no: LIFE09 ENV FI 572

Streets et al. (2003) with the global gridded temperature fields from the CRU3.0 archive[8] of monthly mean temperatures (Brohan et al., 2006). The shares were developed for six years (2000-2006) and an average was eventually used as a representative monthly fraction. Fig. S1 in SI compares this pattern with other existing estimates for selected countries. The importance of considering the temporal distribution of residential combustion emissions developed within ECLIPSE has been demonstrated in Stohl et al. (2013) for the Arctic.

For the energy sector, country-specific monthly patterns were created for selected regions based on available data; for Europe and Russia such data were originally developed in the GENEMIS project (Ebel et al., 1997) and are readily available in the EMEP database; for North America we used the US-EPA Clearinghouse for Emission Inventories (http://www.epa.gov/ttnchie1/emch/temporal/) and the US Energy Information Agency Monthly Energy Review (http://www.eia.gov/totalenergy/data/monthly/); for Thailand the information provided by (Vongmahadlek et al., 2008, 2009) was applied. For all other regions, the temporal distribution file includes constant emissions across the year.

The emissions from open burning of agricultural residues are seasonal since the activity is related to growing cycles and harvesting of different crop types. A global spatial and temporal representation was developed based on the timing and location of active fires on agricultural land in the Global Fire Database GFEDv3.1 (http://www.globalfiredata.org/Data/index.html) combined with annual emissions from GAINS. All active grid cells ($0.5^o$ x $0.5^o$) in the monthly data from 1997 to 2010 in GFED were summed up and normalized. Also for other agricultural activities several patterns were developed but they are more relevant for ammonia and methane emissions and therefore discussed in Klimont et al. (in preparation).

## 3 Emission sources – activity data and emission factors

Here we highlight the contribution of key sources to total emissions and document the sources of activity data and emission factors used in the GAINS model for all relevant sources of particulate matter (PM) emissions, including discussion of differences between several published ECLIPSE datasets. The technology splits and air pollution legislation ~~is~~ are discussed in section 2.2 and 2.3.

The basic statistical data for energy consumption, industrial output, and agriculture originates from International Energy Agency (IEA, 2015a, 2015b), Eurostat (EUROSTAT, 2011), UN Food and Agriculture Organization (http://faostat.fao.org), and several national sources that have been used in the course of collaboration with several partners in Europe (e.g., Amann et al., 2012, 2015) and Asia (e.g., Amann et al., 2008; Purohit et al., 2010; Zhang et al., 2006; Zhao et al., 2013). For several sectors more specific regional data were used; see the discussion in the following source-specific sections. There are also differences in data used for various versions of the ECLIPSE dataset; an overview is provided in Table 1. For activity data, the most significant changes are due to updates of the historical data in versions *V5* and *V5a* where all IEA statistical data ~~was~~ were imported at national level and processed for use in GAINS. Furthermore, for Europe the consultations with

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[8] http://badc.nerc.ac.uk/data/cru/

national experts during the National Emission Ceiling Directive (NEC) revision process led to a number of updates (including activity, emission factors, penetration of control technologies) for the EU-28, specifically in *V4a* (Amann et al., 2012) and then in the *V5a* (Amann et al., 2015) version. Both of these updates were most significant for the year 2010 as new information became available.

The GAINS model database has been developed for five-year periods starting in 1990 and extending to 2050 and, as shown in Table 1, different ECLIPSE versions include estimates for either the whole time horizon or selected five-year periods. There is one exception; in the *V3* dataset we also estimated ~~also~~ global emissions for 2008 and 2009. In order to calculate emission fields for 2008 and 2009 we have used a number of additional sources of information to develop scaling factors for emissions of the year 2005. The exercise was performed at the finest possible sectoral resolution compatible with GAINS but for some regions only key aggregated sectors (see Table 5) were estimated. For most sectors, ~~the country~~ country-specific emission ratios were developed using officially reported emissions ~~by~~ from US-EPA (http://www.epa.gov), Environment Canada (http://www.ec.gc.ca/inrp-npri/), within the UNECE LRTAP Convention (http://www.ceip.at), and 2012 UNFCCC national inventory submissions (http://unfccc.int/). For countries where we found no submissions, emissions for key sectors (Table 5) were linearly interpolated between 2005 and 2010. Additionally, for flaring in the oil and gas industry the emissions for 2008 and 2009 were calculated using GAINS methodology and data on activities available from the NASA report (Elvidge et al., 2011). Finally, for open biomass burning we have used ~~the~~ data from the GFED v3.1 global database (http://www.globalfiredata.org/).

*What is not included and where to find it*

None of the ECLIPSE datasets includes estimated emissions from forest and savannah fires (note that emissions from open burning of agricultural residue are included; see section 3.7), which can be obtained from the GFED v3.1 global database (van der Werf et al., 2010) or a more recent version GFED v4 that was made available subsequently (Randerson et al., 2015). GFED contains emissions for BC, OC, $PM_{2.5}$, and total particle matter (TPM) for the period 1997-2014 in varying temporal and spatial distribution (including gridded dataset) depending on the version (http://www.globalfiredata.org/).

None of the ECLIPSE datasets includes emissions from international aviation but these can be acquired from the Representative Concentration Pathways (RCP) database available at e.g., http://tntcat.iiasa.ac.at:8787/RcpDb/. The data originate~~s~~ from a study by Lee et al. (2009) and were used in the development of the RCPs (Van Vuuren et al., 2011). However, only emissions of black carbon (BC) are included.

Versions *V3* and *V4a* do not include emissions from international shipping and at the time we recommended using datasets developed for the RCP process (Buhaug et al., 2009; Eyring et al., 2010). Version *V5* and *V5a* include international shipping estimates for all PM species (the RCP set contains only BC and OC), which we have developed drawing on the QUANTIFY[9] project spatial distribution (Endresen et al., 2007) and activity data from Buhaug et al. (2009); for more details

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[9] QUANTIFY - Quantifying the Climate Impact of Global and European Transport Systems; European Union Sixth Framework project (https://www.pa.op.dlr.de/quantify/)

see section 3.4.2. The datasets for international shipping, aviation, and open burning have been extracted for use in the ECLIPSE project and can be downloaded (upon request) from the project website http://eclipse.nilu.no.

### 3.1 Residential sector

Several previous studies (e.g., Bond et al., 2004; Cofala et al., 2007; Kupiainen and Klimont, 2007; Lu et al., 2011; Venkataraman et al., 2005) showed that the residential sector is an important source of PM emissions at a regional and global level, especially of carbonaceous species. GAINS distinguishes a number of source categories for residential sector heating and cooking, i.e., fireplaces, stoves, single house boilers and ~~medium~~ medium-sized boilers as well as a number of solid fuels, i.e., fuelwood, agricultural residues, dung, and coal, as well as liquid and gaseous fuels, i.e. kerosene, fuel oil, LPG, and natural gas; see Table 2. The data about fuel consumption used in the GAINS model originate~~s~~ primarily from IEA statistics but ~~is~~ are enriched with additional data from regional statistics and studies. This includes regional, rather than national, statistics of coal use in China (Zhao et al., 2013) but most of all additional assessments of biomass use for cooking and heating in several regions; for US, Canada, Finland, Sweden, and Norway drawing on the collaboration within the Arctic Council (AMAP, 2015); for Australia and New Zealand (Driscoll et al., 2000; Scott, 2005); Asia (Amann et al., 2008; Klimont et al., 2009; Purohit et al., 2010; Venkataraman et al., 2010); and finally for Europe where exchange with national experts led to consideration of several local datasets in the GAINS model (Amann et al., 2015). The data used in the last version of ECLIPSE (*V5a*) for Europe are comparable with the independent fuel estimate by Denier van der Gon et al. (2015). Beyond the total fuel use, the split by fuel and installation types is of high relevance (see discussion in section 2.2).

The global fuel use for cooking and heating used in GAINS ranges from about 2100±200 Tg in 1990 to 2600±200 Tg in 2010 and compares well with the total fuel demand estimated in other global studies; for example, Fernandes et al. (2007) estimated total biofuel use in 2000 at 2460 Tg, which compares with GAINS value of 2200-2500 Tg (the range given owing to uncertainties in assumptions about heat value of various biofuels).

The emission factors aim to reflect real world emissions (e.g., MacCarty et al., 2007; Roden et al., 2006, 2009), i.e., incorporate emission measurements of diluted samples, and have been recently compared and updated for Europe (Boman et al., 2011; Pettersson et al., 2011; Schmidl et al., 2011; Tissari et al., 2008, 2009), specifically for modern stoves and boilers, Asia (Cao et al., 2006; Chen et al., 2009; Habib et al., 2008; Li et al., 2009; Parashar et al., 2005; Venkataraman et al., 2005; Zhi et al., 2008, 2009), and Latin America (Johnson et al., 2008).

Emission factors and shares of BC and OC in particulate mass emissions from selected measurement literature, together with the range of values used in the GAINS model, are presented in Tables S2.1 – S2.4 in the SI (S2), where ~~also~~ a brief characterization of stove and boiler categories used in GAINS is also provided.

### 3.2 Kerosene lamps

Most of the previous emission studies did not highlight particulate matter emissions from kerosene used for lighting, primarily because the information about emission factors and fuel use was either not available or sparse. Only after Lam et al. (2012) reported very high black carbon emission factors, indicating that this is potentially an important 'missing' source, has more work been done to distinguish between kerosene used for cooking and lighting; the new estimates suggest this source might contribute 5-10% of global BC emissions.

Approximately 250 million households (about 1.3 to 1.5 billion people, mostly in developing Asia and Sub-Saharan Africa) lacked access to reliable electricity to meet basic lighting needs in 2010 (IEA, 2012b). These households often rely on fuel-based lighting, with the majority burning kerosene in wick-type lamps (Lam et al., 2012; Mills, 2005); their consumption was estimated at up to 25 billion litres of kerosene per year (Lam et al., 2012). Growing evidence suggests that these light sources pose risks to health (Pokhrel et al., 2010) and the environment (Lam et al., 2012), and improvements to lighting may provide numerous welfare benefits to households (Jacobson et al., 2013).

Annual kerosene consumption ($K_i$) for lighting in GAINS region *i* in year *y* was estimated by using the following expression

$$K_{i,y} = \left(\frac{POP_{i,y}}{HS_{i,y}}\right)\left(1 - ele_{i,y}\right) * 365 \sum_{j=1}^{n}\left(N_{i,j,y} h_{i,j,y} CV_k f_{i,j,y} SC_j\right), \qquad (4)$$

where, *POP* represents population, *HS* household size, *ele* electrification rate, *f* share of device type *j* (either wick lamps or hurricane lanterns), *N* number of kerosene lamps, *h* daily operating hours, *SC* specific kerosene consumption of a device, and $CV_k$ the calorific value of kerosene.

The population data originate~~s~~ from (IEA, 2012a), household size from (UN-Habitat, 2005), the electrification rates from OECD/IEA sources (IEA, 2007, 2011, 2012b) and national data/reports (ESMAP, 2005; GOI, 2011; NSSO, 2007). For India, information about the share of lighting devices (i.e., wick lamps, hurricane lanterns), operating hours~~,~~ and specific kerosene consumption are derived from regional studies (Desai et al., 2010; Mahapatra et al., 2009; Purohit and Michaelowa, 2008). Reported specific kerosene consumption in kerosene lamps varied from 0.005 to 0.042 litre per hour (e.g., Mills, 2003; Pode, 2010) and we assumed 0.006 and 0.02 litre per hour for wick lamps and hurricane lanterns, respectively. Further, we assumed that each household will use three lamps for 6 hours per day, whereas the share of hurricane lanterns is 20 percent for South Asia and 50 percent for other regions.

In India, over 44 percent of rural and about seven percent of urban households reported kerosene as their primary source of lighting in 2004–2005 (NSSO, 2007), and in the lowest four socioeconomic deciles, 60 percent of households use kerosene for lighting (Parikh, 2010). In several of the most populated African countries, including Uganda, Ethiopia, and Kenya, more than 60 percent of the population relies on kerosene as the primary lighting fuel (Apple et al., 2010; IFC/WB, 2008; UBOS, 2010).

Less is known of the quantity of kerosene used for lighting, since it is often difficult to differentiate kerosene used for lighting from that used for other purposes, particularly cooking. The India Human Development Survey 2005 (Desai et al., 2010) results indicate that kerosene lighting accounts for approximately 65 percent (or 5-6 Tg year$^{-1}$) of residential kerosene consumption in India. Lam et al. (2014) observed that use of kerosene for lighting in electrified homes is substantial (due to intermittent and unreliable electricity supply), constituting an approximately equal share of demand as non-electrified households.

Particulate matter emission factors for kerosene lamps used in this work were derived from Lam et al. (2012). The $PM_{2.5}$ emission factor for kerosene lighting (1.92 g $GJ^{-1}$) is approximately 13 times higher compared to that for kerosene used for cooking (0.15 g $GJ^{-1}$), whereas the OC emission factor for kerosene lighting is roughly one third of the kerosene stove. Furthermore, particulate emissions from kerosene lamps are mostly BC (~92%) (Lam et al., 2016).

### 3.3 Diesel generators

At a global scale, diesel generator (DG) sets are not a large source of pollution but locally, and especially in the developing world, they could be responsible for a significant share of air pollutant emissions, especially nitrogen oxides and black carbon. DG sets are the prevailing option for backup power in facilities where continuous power is essential, based on their combination of reliability, durability, affordability, and overall efficiency (Shah et al., 2006). While increasing power deficit and instabilities in the electricity market resulted in rapid growth of the DG set market in several developing regions, DG have been in use all over the world as backup power facilities, primary electricity generation sources in small remote areas or at initial development stage of industrial plants, for irrigation purposes, etc. The DG sets range from small engines to large generators, are operated on very variable fuel quality, and the emission limit values have been typically lagging behind those for mobile engines.

There ~~is~~ are no direct statistical data on fuel use in DG sets as their consumption is typically part of the energy use reported within power plants, commercial, and, potentially, the agricultural sector. Therefore, fuel consumption was estimated from data on number and size of diesel generators as well as regional studies. The resulting fuel use was compared to the IEA statistics for the power and commercial sector and adjusted if necessary so that the overall energy use is consistent with the IEA.

According to a market review in India, annual DG sales in 2010 were about 150,000 units and they are likely to grow at a rate of about seven percent (Frost & Sullivan, 2010) driven by chronic power shortages and prolific growth in industries, infrastructure, telecommunication, information technology (IT), and IT enabled services. The DG market spans from small (15 – 75 kVA) to large (375.1 – 2000 kVA) sets with an estimated diesel consumption of about 5 to 6 billion litres between 2008 (Anand, 2012) and 2010[10]. This represents about 8-9%[11] of total diesel consumption (Anand, 2012; NIELSEN, 2013) and in peak periods up to 18% or even more in some regions (NIELSEN, 2013). In Nepal, electricity deficit has been estimated recently at almost 50% (NEA, 2012), massively increasing dependency on diesel generators. The share of diesel used for DG sets in Nepal is estimated at 15 percent for 2010 (World Bank, 2014a). In Nigeria, total electricity demand is estimated at between 8,000 and 10,000 MW while supply from the national grid is about 4,500 MW, which results in very heavy reliance on DG sets operating most times between 15 – 18 hours a day (Triple E., 2013; World Bank, 2014b). For South Asia (except Nepal), Cambodia, Indonesia and Myanmar we have used the Indian share of diesel consumption in DG sets, whereas in other developing countries~~,~~ the share of diesel use for DG sets is assumed to be one fourth of the Indian

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[10] http://ppac.org.in/

[11] http://www.nipfp.org.in/newweb/sites/default/files/Diesel%20Price%20Reform.pdf

share due to high electrification rates and relatively low power deficit. For sub-Saharan Africa, due to very high power deficit (up to 50 percent), in some regions we have used the share of diesel use in DG sets from Nepal (World Bank, 2014a). For South Korea, diesel consumption in DG sets was less than 0.2 ~~percent~~ % of total diesel consumption (KEEI, 2011). In EU-28, the share of diesel consumption in DG sets is less than 0.4% of the total diesel consumption; however, the share of heavy fuel oil (HFO) use in DG sets is more than 3% of the total HFO used in the EU. Similarly, in United States and Japan the share of diesel consumption is small while the share of HFO is approximately 0.5% and 2%, respectively.

Stationary DG sets are frequently operated in harsh conditions and, until recently, were rarely subject to emission regulation. Information on DG set emissions factors is fairly limited and not necessarily representative for all regions. GAINS model emission factors were developed on the basis of data reported in a number of studies (Anayochukwu et al., 2013; Corbett and Koehler, 2003; Gilmore et al., 2006; Lee et al., 2011; Lin et al., 2008; Shah et al., 2004, 2006; Shi et al., 2006; Tsai et al., 2010; Uma et al., 2004; US EPA, 1996). While it is possible to achieve emissions reductions from diesel combustion through engine modifications and post-combustion measures, we assume that in the period 1990-2010 DG sets operating in the developing world lack any such controls. In case new information will become available, and for future implementation of respective policies, the GAINS model includes a number of post-combustion control technologies such as diesel particulate filters (DPFs), diesel oxidation catalysts (DOC), and fuel-borne catalysts (FBC) offering reduction of gaseous and particulate emissions (Herzog, 2002; Yelverton et al., 2016). Shah et al. (2007) observed that DOC and DOC+FBC technologies were effective in reducing mainly organic carbon (OC) emissions (56-77%), while DPFs showed excellent performance in reducing both elemental carbon (EC) and OC emissions (>90%). The emission factors and shares of BC and OC in particulate mass emissions from measurement literature, together with the range of values used in the GAINS model, are presented in Table S3.1 in SI.

### 3.4 Transport

Globally, the transport sector, including international shipping, is estimated to contribute about 10% of total anthropogenic $PM_{10}$ and $PM_{2.5}$ emissions and up to 25% of BC (Table 8). At a regional level, the role of transport in BC emissions varies strongly and, for example, in Europe and North America was estimated at over 60% in 1996 (Bond et al., 2004) and about 50% in 2005 (Kupiainen and Klimont, 2007) and 2010 in this study, while for East Asia its share grew from about 8 to 23% between 1990 and 2010 (this study). The key source of PM emissions in the transport sector is exhaust emissions from diesel engines with typically light- and heavy-duty trucks playing the largest role; Europe is an exception as policies favouring diesel fuels, in terms of both tax rates and emission limits, resulted in a large ~~a~~ share of diesel cars (Cames and Helmers, 2013). Non-exhaust emissions (brake, tyre, and road wear) represent a relatively small share, especially for carbonaceous particles, but their importance grows over time owing to ever more stringent exhaust emission limits.

The overall energy consumption in the transport sector was taken from Eurostat (EUROSTAT, 2011) statistics for the 28 European Union (EU) member states and from the International Energy Agency (IEA, 2015a, 2015b) for all other countries. Fuel consumption of road vehicles is allocated to the different vehicle types through triangulation with data on the active fleet, their average annual mileage, and their average fuel efficiency. The IEA statistics provide fuel consumption figures separately for rail, aviation, and domestic shipping, however, not for mobile machinery used in agriculture, forestry, industry, and construction and mining sectors. Unless national information is available, as is the case for European countries, the US and Canada, we re-allocate 80% of diesel fuel consumption from the IEA categories “industry” and “agriculture” to construction and agricultural machinery, respectively. International shipping and aviation are not included in the GAINS model but were estimated for the ECLIPSE project separately; see section 3.4.2.

There is a vast literature on PM measurements of internal combustion engines used in road vehicles in both developing and developed countries, including also pre-regulation vehicles (e.g., Cadle et al., 2009; Cheung et al., 2009; Geller et al., 2006; Kirchstetter et al., 1999; Liu et al., 2009; Subramanian et al., 2009; Yanowitz et al., 2000). For all world regions we assume that a certain fraction of vehicles is badly maintained  (e.g., Mancilla et al., 2012), or their emission controls tampered with, which is reflected as the share of so-called high-emitters (McClintock, 1999, 2007; Smit and Bluett, 2011; Yan et al., 2011, 2014); see further discussion in section 3.4.1. For Europe and the USA we draw the emission factors for road vehicles from established emission factor models where experts already synthesized the information (HBEFA 3.1, 2010; Ntziachristos et al., 2009; US-EPA OTAQ, 2011). These emission factors are adjusted to conditions in other world regions.

Kupiainen and Klimont (2004, 2007), Bond et al. (2004), Maricq (2007) are examples of studies which summarized and compared emission factors for various vehicle categories. Most of exhaust PM is emitted in a submicron range, actually within 100 nm, and diesel vehicles typically emit several times more (mass-based) PM than equivalent gasoline engines (e.g., Maricq, 2007); exceptions are old vehicles running on leaded gasoline and pre-regulation 2-stroke mopeds (Klimont et al., 2002b; Kupiainen and Klimont, 2004), while the latest gasoline direct injection engines have PM mass emissions comparable to or even higher than the latest diesel engines with particle filter. It is important to highlight that properly functioning particulate filters reduce PM emissions significantly ~~–~~and, consequently, the absolute level of the latest diesel vehicles is about two orders of magnitude lower than for older generations. The carbonaceous particles represent the largest share with the elemental carbon fraction higher for diesel (50–70%) than for gasoline vehicles (30–40%) (e.g., Kupiainen and Klimont, 2007; Maricq, 2007). Non-exhaust emissions, i.e., brake and tyre wear as well as road abrasion, were updated based on (Denier van der Gon et al., 2013; EEA, 2013; Harrison et al., 2012). Recent roadside measurements showed that tyre wear produces essentially coarse particles, with only a small contribution (<0.5%) in the $PM_{2.5}$ size fraction (Stein et al., 2012). Road abrasion emissions significantly increase when studded tyres are used, ~~e.g.,~~ a common practice in Scandinavian and some Baltic countries. Higher abrasion during winter and spring conditions, average usage period, and application shares are factored into the average abrasion emission factor for the Nordic countries (Kupiainen et al., 2005; Kupiainen and Pirjola, 2011).

PM emission factors for the diverse non-road mobile machinery are much less well established, and only seldom available for developing countries. Moreover, most measurements refer to the mandatory duty cycles rather than real-life operating conditions. For Europe and North America we use emission factors based on (EEA, 2013; OTAQ, 2004; Schäffeler and

Keller, 2008) and transfer to other world regions assuming that technology performs similarly and ~~the~~ under comparable operating conditions.

The contribution from diesel engines used in agriculture, construction, mining, rail, shipping, and as back-up generators has been increasing, not least because the emission legislation lags behind that for road transport, but has been receiving more attention recently (e.g., Kholod et al., 2016)~~(e.g., Kholod et al., 2016)~~. Diesel generators and shipping are discussed in separate sections (3.3 and 3.4.2~~),~~ ); more recent emission factors for diesel locomotives (e.g., Johnson et al., 2013; Tang et al., 2015) are compared with GAINS in Table S4.3, and emission factors for other non-road machinery used in GAINS were summarized earlier (Klimont et al., 2002b; Kupiainen and Klimont, 2004, 2007) and are also included in the supplementary information (SI). Emission factors for key diesel and gasoline engines in the transport sector from recent literature and the GAINS model are compared in Tables S4.1 to S4.5.

### 3.4.1 High-emitting vehicles

On-road remote sensing measurements of vehicles suggest that a relatively small fraction of the fleet is responsible for a relatively large fraction of emissions (e.g., Ban-Weiss et al., 2009; Cadle et al., 1997; Mazzoleni et al., 2004; Subramanian et al., 2009). In the literature, these vehicles have been referred to as: high emitters or high-emitting vehicles, heavy emitters, super emitters, gross emitters, excess emitters or smokers, but in principle highlighting the same problem (Shafizadeh et al., 2004). Reasons for their poor emission performance are variable and can be traced back to malfunctioning or totally inoperative emission controls, low combustion efficiency of the engine, engine oil that is entering the combustion chamber, and/or leakage in the exhaust system between the engine and the emissions control devices (Jimenez et al., 2000; Mazzoleni et al., 2004; Norris, 2001). The shares of high emitters and their contribution to total fleet emissions are variable across countries, with, for instance, only limited evidence in Europe for ~~light~~ light-duty vehicles (Borken-Kleefeld and Chen, 2015; Chen and Borken-Kleefeld, 2016), and more modern vehicles seem to have more durable emission controls (McClintock, 2007). Though there is no doubt in the existence of ~~high~~ high-emitting vehicles, quantifying their emissions is much more speculative.

According to Shafizadeh et al. (2004) two general definitions of high emitters can be identified from the literature: a group of vehicles that (i) account for a certain fraction, e.g., 50 percent, of air pollutant emissions, or (ii) have emissions above a certain emission threshold or cut-off. The GAINS estimation of high emitter emissions is based on the second general definition. The calculation requires two sets of information: (i) the amplification factor which is the ratio between the high and normal emitter emission factors, and (ii) the share of high emitters in the whole vehicle fleet.

The technology-specific amplification factors, i.e., for Euro 1 to 6, were developed based on existing studies mainly from the United States (Ban-Weiss et al., 2009; Durbin et al., 1999; Hsu and Mullen, 2007; Yanowitz et al., 2000) and Europe (Carslaw et al., 2011; Ekström et al., 2004), studying the 90-95th percentile as the cut-off between high and normal emitting behaviour. Similar datasets from Australia (Smit and Bluett, 2011), China (Guo et al., 2007), Thailand (Subramanian et al., 2009) and Mexico City (Jiang et al., 2005) were also studied in order to find which percentiles would represent the local fleets if the amplification factors identified, based on the 90-95th percentiles in the European and US studies, would be applied also there. The identified percentiles then determined what share of the vehicle fleet corresponded to the amplification factors specified for the ~~high~~ high-emitting vehicles. A global coverage of the parameterization was developed using the available studies and databases listed above as benchmarks representative for larger groups of countries and regions. We acknowledge that this definition of the ~~high~~ high-emitting vehicle class is based on a statistical analysis only and currently does not have a technical definition. However, the motivation of the exercise is to single out a portion of the vehicle fleet that might emit significantly more than the majority of the fleet and study the potential importance of such vehicles in total emissions. The amplification factors determined from the studies varied between pollutants, vehicle types and fuels. Table 6 demonstrates the derived amplification factors for light- and ~~heavy~~ heavy-duty on-road vehicles that apply for all countries and all PM species, following the observations reported by Subramanian et al. (2009). We have noted the results by Lawson (2010) who showed that the OC/BC ratio might be different for high emitters than for normal vehicles but have not introduced variable ratios for individual vehicle categories.

The default assumptions about the high-emitter shares are: about 5% for the EU-28, Japan, and Korea; 8% for Australia, Canada, and US; 5-10% for non-EU Europe, 12% for China (except some key cities with a more modern fleet where 10% is assumed); 15% for India, and 20% for other developing Asia, Africa~~,~~ and Latin America. These assumptions are compatible with those used in other global studies (e.g., Bond et al., 2004, 2007, Yan et al., 2011, 2014). In addition, we factor in that the durability of the emission controls ~~have~~ has increased. Therefore, we assume that failure rates decline for the more modern technologies, i.e., above the equivalent of Euro 4, which translates to halving the percentage of high emitters for such vehicles. For example, for Europe or Japan for most recent years this results in a lower overall rate of about 2%, which is consistent with assessments for the US and Europe (Chen and Borken-Kleefeld, 2014; McClintock, 2007).

### 3.4.2 International shipping and aviation

Particulate matter emissions from international shipping contribute about 3-4% of the global total, and while, unlike for $SO_2$ and $NO_x$, this is a rather small share, it is also comparable to the contribution of road transport (e.g., Lack et al., 2009). Aviation contributes only a very small proportion of global PM emissions, e.g., for black carbon its share was estimated at about 0.1-0.2% (Lee et al., 2009; Stettler et al., 2013), of which about 14% were during landing and take-off (LTO) (Stettler et al., 2013).

The GAINS model does not include emissions from these sources and the gridded ECLIPSE datasets *V3* and *V4a* refer to other sources, e.g., datasets developed for the RCP process (Buhaug et al., 2009; Eyring et al., 2010; Lee et al., 2009). However, the more recent ECLIPSE sets (*V5* and *V5a*) include international shipping estimates developed using activity data from Buhaug et al. (2009); fuel consumption data for 2007 were extrapolated to 2010 using GDP. Our extrapolation for 2010 produced fuel consumption similar to the average estimated for the period 2007-2012 (Smith et al., 2015) but larger by about 10% than the IMO estimate for 2010 (Smith et al., 2015). Emissions are estimated for all PM species (the RCP set contains only BC and OC) using emission factors shown in Fig. 5 and spatially distributed drawing on the QUANTIFY project[12], i.e., based on global ship traffic data (Endresen et al., 2007). The fuel consumption data include~~s~~ assumptions about region-specific regulation with respect to fuel quality, i.e., sulphur content of fuels.

The shipping PM emissions and their chemical, physical, and optical properties have been analysed for various types of fuels, engines, and vessels, as well as operating conditions, e.g., load factors (Agrawal et al., 2008, 2010, Lack et al., 2008, 2009; Moldanova et al., 2009; Murphy et al., 2009; Petzold et al., 2008, 2010). Further studies reviewed and compared emission factors (Buhaug et al., 2009; Dalsoren et al., 2009; Lack and Corbett, 2012). The particulate matter emission profile, including BC and OC, presented in Fig. 5, was developed on the basis of the studies listed above.

### 3.5 Large scale combustion

Solid fuel combustion in large boilers used in power plants and industry has been a major source of primary particulate matter emissions and although efficient reduction technology exists and is typically required by law, about 15% of total global anthropogenic $PM_{2.5}$ emissions in 2010 originated from this source. At the same time, since the 1990's emissions declined by over 30% and its share dropped from over 20% to 15%. Primary PM from combustion can be divided into two major categories: (i) ash, formed from non-combustible mineral constituents in fuel, which vary from a few to over 30% depending on fuel quality, and (ii) carbonaceous particles, e.g., char, coke and soot, which are formed by pyrolysis of unburned fuel molecules (e.g., Seinfeld and Pandis, 2012). The largest particles remain in the boiler and are removed with bottom ash, while smaller (typically <100 μm) are entrained in combustion gas forming fly ash. Emissions of elemental and organic carbon from such installations are small due to the high combustion temperature, oxidizing conditions, and long residence times (e.g., Ohlström et al., 2000); only about 2% of global total black carbon was estimated to originate from this source (Bond et al., 2004, 2013; Cofala et al., 2007).

The principal statistical data for energy use in the power sector and industry used in GAINS originates from International Energy Agency (IEA, 2015a, 2015b), Eurostat (EUROSTAT, 2011), and national sources, especially for Europe (e.g., Amann et al., 2012, 2015) and Asia (e.g., Amann et al., 2008; Purohit et al., 2010; Zhang et al., 2006; Zhao et al., 2013). The national sources and consultations were especially useful to distribute fuel use among different types of plants; see discussion in section 2.2.3.

The PM emission factors in GAINS are calculated considering region-specific fuel properties (heat value, ash content), installation-specific parameters (ash retention in boiler, size distribution), size-specific efficiency of control equipment (cyclones, wet scrubbers, electrostatic precipitators, fabric filters); see Eq. 1, Eq. 2, and discussion in section 2.1. Detailed review of measurement studies, methodology and assumptions applied in GAINS has been documented in a number of earlier reports and papers (Klimont et al., 2002b, 2009, Kupiainen and Klimont, 2004, 2007; Zhang et al., 2006; Zhao et al., 2013). Key updates with respect to emission factors have been done for Europe within the work for the European

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[12] https://www.pa.op.dlr.de/quantify/

Commission (Amann et al., 2015) and China, where the latest information about efficiency and penetration of control measures was used (Zhao et al., 2013).

### 3.6 Industry

There are many industrial processes that emit particulate matter to the atmosphere and the origin of these emissions is often more complex than that of stationary combustion since there are several process stages, fugitive sources, and the process designs vary significantly across the world. The particular processes will also differ with respect to emission characteristics, i.e., PM size distribution and chemical speciation. The GAINS model distinguishes tens of industrial processes, including several within the iron and steel sector, non-ferrous metals, cement and lime, petroleum refining, coal mining, gas flaring, and production of bricks, coal briquettes, mineral fertilizers, glass, carbon black, and pulp. Extensive discussion of these sources, including their particulate matter and carbonaceous aerosols emissions and mitigation measures in GAINS is available from previously published reports (Klimont et al., 2002b; Kupiainen and Klimont, 2004). The estimates presented in this paper rely for most sectors on the characteristics presented in those reports, however with updated emission factors for a number of regions and specifically a new structure ~~of~~ for the three sectors most~~ly~~ relevant for carbonaceous particles, i.e., coke ovens, brick making, and gas flaring.

While there are ~~well~~ well-established PM control technologies applicable to most of the sources (Klimont et al., 2002b; Kupiainen and Klimont, 2004; Maithel et al., 2012) and typically, even in the developing world, there exists legislation prescribing emission limit values, this sector remains among the most uncertain in terms of emission estimation of total PM as well as carbonaceous aerosols. We estimate that, at a global scale, industrial processes contributed between about 13 and 20% of $PM_{2.5}$ emissions in 1990 and 2010 and total emissions grew in this period by over 60%. Regional shares might be much larger, e.g., for China this share was~~ere~~ estimated at over 30% in 2010 and grew by nearly a factor of three compared to 2000, or significantly lower, e.g., for Africa less than 5%. For most regions, key $PM_{2.5}$ sectors include cement and iron and steel production, representing globally about 75% of industrial emissions of $PM_{2.5}$. For carbonaceous particles, this sector plays a slightly less important role from the global perspective; Bond et al. (2004) estimated its contribution at about 13% to BC emissions, primarily from coking and brick making. This is broadly consistent with our assessment, although we estimate a somewhat lower share of about 10% globally, of which about a third comes from gas flaring, and there is very strong regional variation from less than ~~a~~ one percent to over 20%, especially in regions with high oil production, e.g., Middle East, Russia.

The principal statistical data used in GAINS originates from international sources (Elvidge et al., 2009; EUROSTAT, 2011; IEA, 2015a, 2015b), and national sources, especially for Europe (e.g., Amann et al., 2012, 2015) and Asia (e.g., Amann et al., 2008; Heierli and Maithel, 2008; Huo et al., 2012; Purohit et al., 2010; Zhang et al., 2006; Zhao et al., 2013).

The PM emission factors used in the GAINS model have been discussed in previously published reports (Klimont et al., 2002b; Kupiainen and Klimont, 2004) and key updates concern the region-specific primary technology allocation and implementation rates of control technologies – as discussed in sections 2.2.4 and 2.3. For coke manufacturing (see 3.6.1), brick production (see 3.6.2), and gas flaring in the oil and gas industry (see 3.6.3) more significant changes were introduced with new technology and region-specific emission factors.

### 3.6.1 Coke production

Total coke production grew by about a factor of two in the 1990-2010 period and most of the change took place after 2000 when China increased ~~their~~ its production by about a factor of four from just over ~~a~~ 100 Tg to about 400 Tg coke, which represented over 60% of global production in 2010 (Huo et al., 2012) and see http://www.statista.com. China's coke sector undergoes a significant transformation, moving from low efficiency and high emission indigenous ovens to highly mechanized recovery ovens, following the world trend (Huo et al., 2012; Polenske, 2006). Several of the other producing countries remained fairly constant or reduced their output in the last decade, e.g., US, Europe, former Soviet Union region, and only a few increased their production, e.g., India, but from the global perspective these changes were not very significant (http://www.statista.com).

There are only few measurements of PM emissions from coke plants, and the established emission factors show a wide range. This is partly driven by the varying technology but also owing to the sources of emissions from coke manufacturing since they include several stack and fugitive sources. In the GAINS model, we have constructed a PM emission profile based on the US EPA Compilation of Air Pollutant Emission Factors (AP-42)[13] and SPECIATE[14] (US EPA, 1995, 2002) as discussed in Klimont et al. (2002b) and Kupiainen and Klimont (2004), and updated it with more recent measurements discussed in Huo et al. (2012) and Weitkamp et al. (2005). For uncontrolled ovens, GAINS emission factors for $PM_{2.5}$ range from about 2 to 4.8 kg $t^{-1}$ coke, the upper bound being representative for China and the range reflecting different oven types across the global regions. For BC and OC, the emission factor range is 0.28 – 1.3 kg $t^{-1}$ and 0.46 – 2.2 kg $t^{-1}$, respectively, with upper range values representing Chinese indigenous ovens. The PM emission factors for China are comparable to the ones used in recent Chinese studies (Huo et al., 2012; Lei et al., 2011) and the ratio of BC/OC of about 0.6 is also consistent with the estimates by Weitkamp et al. (2005). Owing to a lack of specific data for various world regions, we assume little change in emissions factors over time for the developing world, although the transition in China reported in Huo et al. (2012) was considered, and for OECD countries the emission factor trend follows reported emissions, where available.

### 3.6.2 Brick kilns

The brick making industry is dominated by production in the developing countries, where over 95% of global output, estimated at about 1.5 trillion bricks per year (e.g., Schmidt, 2013), is produced and most of it in fairly inefficient and polluting kilns. In India, over 70% of kilns, or about 100,000, are clamp kilns, the least efficient kiln that remains widespread in the developing world. More than 1.2 trillion bricks per year are produced in Asia alone, which is associated with the use

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[13] https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emission-factors

[14] SPECIATE is the US EPA repository of volatile organic gas and particulate matter (PM) speciation profiles of air pollution sources: https://www.epa.gov/air-emissions-modeling/speciate-version-45-through-32

of over 100 million tons of coal as well as other fuels including agricultural residues, dung, and waste (Heierli and Maithel, 2008; Schilderman and Mason, 2009). The largest ~~brick~~ brick-producing countries in Asia are China, India, Pakistan, Bangladesh and Vietnam (AIT, 2003; FAO, 1993; Heierli and Maithel, 2008; Maithel, 2014). Worldwide non-automated brick production, including artisanal brick kilns, in developing countries is about 1.25 trillion bricks per annum and is distributed between three main regions (i) China – about 700 billion bricks or 56%, (ii) India – about 150 billion bricks or 12%, (iii) Asia, Africa, South America & Mexico – about 400 billion bricks or 32%. In contrast, worldwide ~~machine~~ machine-made brick production using automated kilns~~,~~ is approximately 125 billion bricks, with Australia's brick production accounting for only 2 billion, UK 4 billion, USA 8 billion, China 100 billion, and other developed countries approximately 11 billion bricks. A summary of the studies used to compile the brick production data is provided in SI (S5) along with the activity data used in ECLIPSE *V5a* for key global regions (Table S5.2).

Even though from the global perspective, the brick manufacturing sector does not represent a major share of particulate matter emissions, about 1-2% for $PM_{2.5}$ according to our estimates and less than 5% for BC (e.g., Bond et al., 2004, 2013), the regional impacts might be much more significant (Guttikunda et al., 2013; Le and Oanh, 2010; Skinder et al., 2014). And while many countries may have emissions standards, i.e., maximum permissible concentrations of several pollutants, including PM, the enforcement is difficult for several reasons including relatively few measurements available. Maithel et al. (2012), Weyant et al. (2014), and Rajarathnam et al. (2014) reported particulate matter measurements for key brick kiln production technologies in Asia (primarily India and Vietnam), and a few studies, focusing on toxics and black carbon, were performed in Mexico (Cardenas et al., 2012; Christian et al., 2010; Maíz, 2012); the latter covered several types of kilns including the Marquez kiln (MK) that is specific to Latin America. For the main brick producing technologies in South Asia, the PM emission factors derived from the above measurements are lower by over 30% for BC and 90% for $PM_{2.5}$ than previously estimated values (Weyant et al., 2014), which were used in several regional (Klimont et al., 2009; Lu et al., 2011; Ohara et al., 2007; Zhang et al., 2009) and global inventories (Bond et al., 2004; Cofala et al., 2007; UNEP/WMO, 2011). Additionally, the BC/TC ratio appears higher than previously thought (Weyant et al., 2014).

The emission factor set used in GAINS to calculate ECLIPSE ~~set~~ values is more in line with the currently available measurements although it was developed prior to the publication of measurements by Weyant et al. (2014); compare Table S5.1 in the SI, where current GAINS emission factors for $PM_{2.5}$, BC, and OC are compared with the previous GAINS dataset and recent measurements by Weyant et al. (2014). Also the EC/TC ratio in GAINS, from about 0.67 for zig-zag, about 0.75 for clamps, downdraft, moving chimney BTK, and 0.93 for fixed chimney BTK, resembles the measurements by Weyant et al. (2014).

### 3.6.3 Gas flaring

Understanding venting, flaring, and associated gas utilization practices in the oil industry has been of high relevance for the assessment of methane emissions while it was not considered as a potentially important source of air pollution. Consequently, non-$CO_2$ emissions from flaring of associated gas in oil industry were not part of previous inventories (e.g., Bond et al., 2013), including the datasets used in the IPCC assessments. We have developed the first global estimate of air pollutant emissions from this activity, including black carbon, which was used first in the studies focusing on the role of black carbon and other short-lived climate forcers in climate mitigation (Bond et al., 2013; Shindell et al., 2012; UNEP, 2011; UNEP/WMO, 2011; World Bank and ICCI, 2013). Within the ECLIPSE project, an update and future mitigation scenarios (Klimont et al., in preparation) were developed and used in several regional and global modelling exercises (Stohl et al., 2013, 2015).

Associated petroleum gas (APG) is gas that is associated with the oil in the reservoir and once oil is extracted, the dissolved gas follows and is commonly separated from the oil and either vented or flared. The volumes and composition of APG depend on several factors including the nature of the oil reservoir, degree of depletion, etc. (PFC Energy, 2007; Røland, 2010). While the APG could be utilized, the lack of developed markets, missing infrastructure, no legislation, etc. resulted in very low recovery rates before 1980; only in the last decades has the flaring trend been decoupled from oil production but the level of gas utilization varies greatly among the producing regions. Globally, about 140-160 billion m$^3$ (bcm) APG have been flared annually, which represents about 5% of 2009 global natural gas consumption or about 30% of European Union demand (Elvidge et al., 2009). Regions where the largest volumes of gas are flared include Middle East, Russia, Northern Africa, Nigeria~~,~~ and Venezuela, representing about 70-80% of the global total (Elvidge et al., 2009, 2013). There are significant uncertainties in estimates of flared volumes as metering is rare and official estimates differ significantly from remote sensing data or even between different official versions, e.g., for Russia governmental sources reported for 2006 about 15-20 bcm of APG flared while Global Gas Flaring Reduction Initiative (GGFR) estimates were about 40-60 bcm (PFC Energy, 2007). The reported share of APG flared in Russia in 2006 varied from 27% (governmental sources) to 75% (NGOs) with 45% estimated by PFC Energy (2007) (Røland, 2010). For Nigeria, flaring volumes have been estimated or reported between 10 to 25 bcm indicating that up to 70% of APG is flared (Aghalino, 2009; Ite and Ibok, 2013). While for several countries APG utilization rates have been increasing (Elvidge et al., 2009; Haugland et al., 2013), Russia made relatively little progress until 2010 in spite of new legislation requiring a 95% recovery rate (Evans et al., 2017; PFC Energy, 2007; Røland, 2010) ~~(PFC Energy, 2007; Røland, 2010)~~. For US, flaring volumes increased by about a factor three between 2006 and 2011 owing to the boom in unconventional gas and oil production (Elvidge et al., 2013). GAINS activity data relies on the time series of gas flaring volumes developed within the GGFR initiative (Elvidge et al., 2007, 2011).

There is a very limited number of measurements of flaring emissions allowing the establishment of a representative set of emission factors where local flare operating conditions and APG properties could be considered. Some of the earlier published PM emission factors (about 2.6 g m$^{-3}$) referred to landfill (CAPP, 2007) or refinery flares (US EPA, 1995) and are generally considered inappropriate. A new technique for quantitatively measuring soot emission rates in flare plumes under field conditions has been reported by the Carlton University group (Johnson et al., 2011) and while their average BC emission factor of 0.51 g m$^{-3}$ (McEwen and Johnson, 2012) considers representative fuel mixtures, their measurements were performed on laboratory-scale flares, which might underestimate real-world emissions. The first ECLIPSE datasets include flaring emissions calculated with one BC emission factor of 1.6 g m$^{-3}$ gas flared, assuming that real-life flares perform much worse than laboratory measurements. In the later ECLIPSE set *V5a*, region-specific PM emission factors were developed considering a more recent study measuring emissions from flares in the Bakken region (Schwarz et al., 2015) which confirmed the order of magnitude measured by McEwen and Johnson (2012) by establishing an upper bound BC emission factor of 0.57±0.14 g m$^{-3}$. We have assumed that such emission rates are representative for ~~well ~~well-operated flares, i.e., ~~OCED ~~OECD countries. For other countries we retained the previously used value of 1.6 g m$^{-3}$ but considered, where available, the composition of flared gas that apart from methane includes several heavier hydrocarbons. The relationship between BC emission factors and heat value of flared gas has been proposed by McEwen and Johnson (2012) and was also applied in estimates for Norway (Aasestad, 2013) and Russia (Huang et al., 2015).

The range of current BC emission factors in GAINS is ~0.5-1.75 g m$^{-3}$, the upper bound represents values for Russia, and the estimated heat value of APG varied from about 41 to 50 MJ m$^{-3}$. Huang et al. (2015) suggested even higher BC emission factors for Russia (2.27 g m$^{-3}$), assuming a local APG composition with estimated heat value of about 75 MJ m$^{-3}$ and extrapolating linearly from the relationship from McEwen and Johnson (2012~~); ~~), but well beyond the range presented there. Finally, the most recent measurements of BC from flaring, also in the Bakken field, estimate much lower overall emission factors of 0.13±0.36 g m$^{-3}$ and characterize flares without visible smoke (Weyant et al., 2016) and therefore, likely not representative for regions with visible high-density smoke, e.g., Russia, Nigeria, Middle East, Northern Africa (e.g., Aghalino, 2009; Elvidge et al., 2013; Pederstad et al., 2015). We assume that all PM from flaring is PM$_{2.5}$ and BC and OC represent about 78% and 16%, respectively. These assumptions are broadly consistent with the results of McEwen and Johnson (2012) who reported a BC/OC share of 80/20 and Fortner et al. (2012) measuring 4-20% of OC and over 95% of PM within PM$_{2.5}$.

### 3.7 Agricultural waste burning

Bond et al. (2004) estimated that globally about 7 and 15% of anthropogenic (excluding forest and savannah fires) BC and OC emissions originated from this source in 1996; our own estimates point to a slightly lower share in carbonaceous particles emissions but mostly because our total, not agricultural burning, estimates are higher. At the same time, for several regions this source might be even more important, e.g., for Brazil we estimate its contribution at up to 15% of PM$_{2.5}$ and 10% of BC emissions. Finally, agricultural burning has a strong seasonal pattern (see also section 2.4.1.) and has also been ~~also ~~linked with heavy smog and haze episodes (e.g., Mukai et al., 2015; Stohl et al., 2007).

Typically assessment of global emissions from open field burning of agricultural residues is based either on a compilation of national reports/sources (e.g., Bond et al., 2004; EC-JRC/PBL, 2010) or on remote sensing data which characterize the magnitude and spatial distribution of open biomass burning including agricultural, savannah, and forest fires ~~(e.g., van der Werf et al., 2010; Wiedinmyer et al., 2011), ~~(e.g., van der Werf et al., 2010; Wiedinmyer et al., 2011); however, it has been shown the latter underestimates small open fires (e.g., Randerson et al., 2012). Niemi (2007) compared various datasets for all open biomass sources and developed the first global activity set for the RAINS model drawing on EDGAR3.2FT2000 (Van Aardenne et al., 2005) which we have further extended and updated to accommodate other data sources, allowing gaps to be filled for several countries. Specifically, we have used estimates from the global studies (Bond et al., 2004), a number of regional estimates (Cao et al., 2008; Oanh et al., 2011; Pettus, 2009), reporting of emissions to EMEP (http://www.ceip.at), and bilateral discussions within the revision of the European air pollution policy (Amann et al., 2015). Our global estimate of open burning of agricultural residue has been fairly constant in the assessment period varying from about 485 to 515 Mt between 1990 and 2010; this estimate is comparable with 475 Mt for 1996 by Bond et al. (2004) and higher than the original EDGAR3.2FT2000 of 252 Mt of residue burned in 2000.

To derive particulate matter emission factors, we have relied on Akagi et al. (2011), Andreae and Merlet (2001), Turn et al. (1997), and Hegg et al. (1997); the latter was used for the OM/OC ratio, which we assumed to be 1.7 as discussed in Kupiainen and Klimont (2004). The default emission factors used in GAINS (all values in g $kg^{-1}$) are 8.5 for TSP, 7.1 for PM10, 6.3 for $PM_{2.5}$, 5.6 for $PM_1$, 2.62 for OC and 0.83 g $kg^{-1}$ for BC. Using data from Turn et al. (1997), these values were adjusted for specific regions considering typical types of crops; for example, for regions with a high share of rice production (primarily Asia) the values of BC and OC factors were estimated at 0.6 and 2.2 g $kg^{-1}$.

## 3.8 Waste

Open burning of solid waste is a widespread method, especially in the developing world, to reduce the volume or odours of dumped or uncollected municipal solid wastes (EAWAG, 2008) and it has been identified as a significant source of particulate matter and hazardous air pollutants to the atmosphere (Christian et al., 2010; Hodzic et al., 2012; Kumar et al., 2015; Wiedinmyer et al., 2014). The estimated magnitude of emissions and contribution to PM concentrations vary widely across the studies, ranging from a few percent to nearly 50% of the total contribution in particular regions. While large uncertainties remain owing to only ~~few~~ scarce measurements and difficulties in finding reliable data on waste collection, recycling, and disposal rates, the open burning of residential waste is a potentially important source of PM, especially in the developing world.

To estimate the region-specific share of the municipal solid waste (MSW) that is burned, we used a mass balance approach described in the IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006b). As a starting point, we used the IPCC reported data on MSW generation and management and assumed that the category "*other MSW management, unspecified*" represents the upper limit for the open burning of residential waste. However, the IPCC values were not used directly in many cases, because the IPCC unspecified fractions are in some cases relatively high, up to 60 percent, and also because not all unspecified mass is necessarily burned. We have additionally used information on percentages of commonly used MSW disposal methods in other studies (CEPMEIP, 2002; EAWAG, 2008; Neurath, 2003); the final fraction of open burning from the total waste produced in the developed world was estimated to vary between 0.5 and 5% and for the developing world the region-specific fractions were estimated at 10-20%. The GAINS model estimate of the global MSW is about 1500 to 2150 Tg in the period 1990 to 2010 of which about 115 to 160 Tg were estimated as openly burned. While the total waste generation rate is consistent with other studies (e.g., Christian et al., 2010; Wiedinmyer et al., 2014), the open burned fraction differs significantly owing to different assumptions about the fraction burned and practices in urban and rural areas. For example Bond et al. (2004) and Wiedinmyer et al. (2014) estimated that 33 and 970 Tg of waste are burned; the latter is still about six times larger than GAINS. We were not able to consider the results of Wiedinmyer et al. (2014) in GAINS yet, but a comparison at the national level shows that GAINS has significantly lower estimates for most of the developing countries as well as Europe; for the latter, GAINS is consistent with national reporting and often a factor 5 to 10 lower than Wiedinmyer et al. (2014). For the US and Canada GAINS has a factor of 2-3 higher estimates (also consistent with the US EPA and Environment Canada).

The PM emission factors used in GAINS were derived from– Akagi et al. (2011) and Christian et al. (2010) and are consistent with the ones used by Wiedinmyer et al. (2014). These are (all in g kg$^{-1}$) 9.5 for $PM_{10}$, 8.74 for $PM_{2.5}$, 6 for $PM_1$, 5.27 for OC, and 0.65 g kg$^{-1}$ for BC.

## 3.9 Other sources

The GAINS model also includes several other sources of PM which at a larger scale represent a rather small contribution but could be of relevance locally. These are mostly non-combustion (fugitive) emission sources and include animal livestock, storage and handling of bulk industrial and agricultural products, arable land related agricultural activities, and construction works. Additionally, emissions from cigarette smoking, barbeques, and fireworks are considered. Note that windblown dust and emissions from unpaved roads are not included (see also introduction to section 3).

The predominant sources of PM from animal housing include feed and faecal material, bedding, skin, hair, mould, and pollen. Size-specific PM emission factors were developed in GAINS drawing on the results of measurements done in Europe (e.g., ICC &SRI, 2000; Louhelainen et al., 1987; Takai et al., 1998) which are discussed in more detail in Klimont et al. (2002b). The values presented in that report were adapted considering region-specific length of the housing period (time animals spend indoors) which is a regional parameter in the model, also relevant for estimation of ammonia emissions. For dairy cows the $PM_{10}$ factors range from 0.22-0.43 kg animal$^{-1}$ per year, for beef 0.11-0.43 kg animal$^{-1}$, for poultry about 0.05 kg animal$^{-1}$, and for pigs 0.4-0.45 kg animal$^{-1}$. The share of $PM_{2.5}$ is about 22% with the exception of pigs where it was estimated at about 17%; no BC or OC emission factors were assumed. Emissions from arable farming include harvesting, ploughing, tilling, etc. The GAINS $PM_{10}$ emission factor varies from 0.8 to 2 kg ha$^{-1}$ and the $PM_{2.5}$ is assumed to represent about 22% of $PM_{10}$. These revised numbers, compared to the earlier GAINS values discussed in Klimont et al. (2002b), draw on the more recent work in Germany and France discussed within the EU air quality consultation (Amann et al., 2015).

Emissions from storage and handling of bulk industrial (coal, iron ore, fertilizers, cement, other) and agricultural products, as well as from construction activities, are estimated using emission factors discussed in Klimont et al. (2002b). For the latter, some updates were made based on national consultations within work on the revision of the EU air quality policy (Amann et al., 2015) and the recent range for $PM_{10}$ is 0.07 – 0.22 Gg per million m$^2$ of constructed floor space, with a share of $PM_{2.5}$ assumed at 12% and no primary carbonaceous particles.

For cigarette smoking we assume a $PM_{2.5}$ emission factor of 0.01 – 0.0165 kg capita$^{-1}$ (equal to $PM_{10}$) and a share of BC and OC as 0.5% and 60%, respectively (Klimont et al., 2002b). Also for barbeques, a per capita emission factor is established, i.e., 0.02 – 0.075 kg capita$^{-1}$ with a share of BC and OC assumed ~~as~~ at about 15% and 50%, respectively (Klimont et al., 2002b). Only very few regional estimates were available for these sources, specifically identified within the discussion in Europe ~~(Amann et al., 2015),~~ (Amann et al., 2015); therefore, for most countries the same emission rates are used.

## 4 Results and discussion

Global, regional and sectoral emissions of particulate matter (PM) distributed into several size bins ($PM_{10}$, $PM_{2.5}$, $PM_1$), as well as into black and organic carbon, are shown in Table 7-8 for 2010 and Fig. 6-7 for the period 1990-2010; Table S6.2-S6.6 in the SI show global emissions of PM species for 25 global regions in the period 1990-2010. To our knowledge, these estimates represent the first global dataset of anthropogenic emissions where size-specific mass PM calculation, including BC and OC, was performed using a uniform and consistent estimation framework. Emissions are also allocated into a 0.5$^o$ x 0.5$^o$ (longitude-latitude) grid and available freely for a number of datasets[15]. Finally, the PM estimates are consistently linked with the emissions of other air pollutants and greenhouse gases for the same time period, as well as their future projections developed with the GAINS model (Klimont et al., in preparation).

Total emissions of particulate matter (including open burning based on GFED3.1 database but excluding windblown dust) in 2010 are estimated at about 111 Tg for $PM_{10}$, 81 Tg for $PM_{2.5}$, 71 Tg for $PM_1$, 9.5 Tg for BC, and 33 Tg of OC. ~~Anthropogenic~~ The anthropogenic contribution dominated all species except OC and OM, i.e., about 55% of $PM_1$, $PM_{2.5}$, and $PM_{10}$, 75% of BC, and 40% for OC and OM (Table 7). For all ~~considered~~ PM species considered, sources in Asia represented over 60% of the global anthropogenic total (Table 7), with residential combustion being the most important sector although its share declines with increasing particle size: about 60% for BC and OC, 45% for $PM_{2.5}$ and less than 40% for $PM_{10}$ for which large combustion sources and industrial processes are equally important (Table 8).

In contrast to several local and regional atmospheric modelling studies, the global modelling community has been relying so far on the assumption that anthropogenic $PM_{2.5}$ emissions are sufficiently well represented by the sum of black carbon and primary organic PM, often referred to as POM. This total fine PM mass has been typically estimated as BC+1.4*OC[16] and only recently have a number of models included more detailed aerosol schemes accounting for varying BC/OC ratios while still largely neglecting the anthropogenic dust component (e.g., Philip et al., 2017). Combining such estimates with windblown dust and open biomass fires to arrive at the total $PM_{2.5}$ might be sufficient from the perspective of global climate impacts of primary PM aerosols; however, the health impacts could be severely underestimated in some regions where the non-carbonaceous share of anthropogenic fine particulate matter is significant (Fig. 6).

We argue that assessment of health impacts due to PM using results of the global emission projections developed in the first place for climate simulations, e.g., Representative Concentration Pathways (RCP), which included anthropogenic BC and OC, windblown dust, and open fires but not the non-carbonaceous component of primary $PM_{2.5}$ and $PM_{10}$ emissions

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[15] http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html

[16] The 1.4 has been the most commonly used OM/OC ratio (Aiken et al., 2008)

originating from combustion, industrial processes, and some fugitive sources, might lead to inconsistent results and underestimation of PM concentrations and regional impacts. This study provides the first global assessment of the role non-carbonaceous particle emissions play in total anthropogenic $PM_1$, $PM_{2.5}$, and $PM_{10}$ mass emissions and could prove more appropriate to use in global modelling studies of health impacts as well as climate. Moreover, while at the global level, the ratio of anthropogenic emissions of $PM_1$ and $PM_{2.5}$ to (BC+POM) is about 1.3 and over 1.6, there are important differences between the regions and the emission ratios have been changing over time (Fig. 6). For example, in 2010 we estimate for Asia an emission ratio of two for $PM_{2.5}$/(BC+POM) while for North America the same ratio is about 1.5 (Fig. 6, Table 7). In Europe, including Russia, this ratio has changed from about three in the early 1990's, where primary PM emissions from poorly controlled coal power plants and heavy industry (not a large source of carbonaceous particles – compare Fig. 7) dominated the total, to below two in 2010 (Fig. 6). Even when the emissions from open biomass burning (forest and savannah fires) are taken into account, and most of these occur far from densely populated areas, the total $PM_{2.5}$ mass emissions are over 20% larger than the BC+POM (Table 7).

We estimate that about 75% of global anthropogenic emissions of $PM_{10}$ are $PM_{2.5}$ and while there was only little change in that ratio (slight increase) in the last decade at the global level, more significant variation has been observed across sectors (Fig. 7). Combustion of liquid fuels, biomass, and waste produces typically over 90% of $PM_{2.5}$ in $PM_{10}$ but for several industrial processes, power and industrial boilers burning coal, and coal production, distribution and storage, emissions of $PM_{2.5}$ represent only 40-60%. Carbonaceous particles (BC+OM) emissions play a key role in $PM_{2.5}$ representing over 60% with the largest contribution from residential combustion (about 80%) and transport and agriculture (each about 10%). Nearly 90% of $PM_{2.5}$ emissions from residential boilers and cooking and heating stoves is BC+OM of which over 20% is BC. ~~Similarly~~ A similarly high share of BC+OM is estimated for the transport sector but it varies between about 95% for road transport and 80% for non-road vehicles~~,~~ ; however, the share of BC is much larger than for residential combustion: 35 – 45% of $PM_{2.5}$ emissions from transport (including non-exhaust) is BC. ~~Few~~ A few of the smaller sources, agricultural residue and trash burning, also have a ~~also~~ large share of BC+OM (over 80%) but rather small contribution of BC. Combustion of solid fossil fuels in power and industrial boilers, as well as most industrial processes (except brick manufacturing in traditional kilns and possibly coke making), are characterized by a very low share of carbonaceous particles (below 5%).

### 4.1 Regional distribution and temporal trends

Total anthropogenic emissions of $PM_{2.5}$ and BC in 2010 have a similar spatial distribution (Fig. 8). Emission densities are generally the highest in Asia, however, there are some important differences in the contributions of various sectors to both species as well as across regions. Residential combustion plays a key role but appears far more important for BC, where it represents nearly 60% of the global total (Table 8) and an even higher share for Asia and Africa; for $PM_{2.5}$ this sector contributes globally about 45%. While for $PM_{2.5}$ the energy and waste sector (incl. agricultural burning) and industry make up most of the remaining emissions (25% and 17.5%, respectively), they represent just over 10% of BC emissions (Table 8 and Fig. 8). Industrial emissions appear much more important in Asia (Fig. 8) and while there are several processes contributing to $PM_{2.5}$ emissions, for BC brick and coke production make the most and represent up to 12% of Asian emissions, globally about 6%. Some sector contribution patterns are similar across continents, for example, for North America, Latin America~~,~~ and Europe transport and the residential sector dominate BC emissions, while for $PM_{2.5}$ it is mostly energy and the waste sector, except Europe where ~~also~~ residential combustion also appears important (Fig. 8). For Africa, residential combustion is the key source of all PM with the exception of a few areas like Republic of South Africa or oil producing countries where the energy sector is an important source. It is particularly striking to see the difference in the source contributions to BC emissions in Africa and Asia where the most important source is the residential sector, but while in Africa other sources are barely visible, for Asia there are important contributions from ~~the~~ transport and industry (Fig. 8). The other feature worth highlighting is the difference in relative importance of the transport sector for $PM_{2.5}$ and BC emissions (about 8% and 24% at ~~a~~ the global level, respectively) which is clearly visible in the third row of maps in Fig.8.

We estimate that global emissions of PM have changed little in the period 1990-2010 showing a strong decoupling from the global increase in energy consumption and, consequently, $CO_2$ emissions (Fig. 6). However, there are very different regional emission trends with a particularly strong increase in East Asia and Africa, and a strong decline in Europe, North America and Pacific. The development of $PM_{10}$ and $PM_{2.5}$ emissions is fairly similar with a slightly faster growth of $PM_{2.5}$ (+8%) than $PM_{10}$ (+4%) at ~~a~~ the global level. The difference is mostly due to reductions of industrial emissions in Europe and Russia following the political and economic transition in Eastern Europe that started already in the mid-80s. This economic restructuring resulted in closure or transformation of inefficient and polluting heavy industries which in turn brought in about 55 and 60% reduction of $PM_{2.5}$ and $PM_{10}$ emissions between 1990 and 2010~~;~~, most of which was achieved before 2000 (Fig. 6). Also, North American and Pacific emissions declined in this period by about 30%. In contrast, $PM_{10}$ and $PM_{2.5}$ emissions in East Asia and Africa increased by about 40-50% and those of Other Asia and Latin America by about 10%. The stark differences in regional trends resulted in important changes in the spatial pattern of PM burden. The European, North American, and Pacific contribution to global emissions dropped from nearly 30% in 1990 to well below 15% in 2010 while Asia's contribution grew from just over 50% to nearly 2/3 of the global total in 2010 (Fig. 6, Table 7, Table S6.2-S6.3).

For black carbon (BC), the regional changes were less dramatic but the global emissions are estimated to grow by about 15% by 2010 compared to 1990, mostly driven by increases in Asia (about 30%) and Africa (over 40%) (Fig. 6, Table 7, Table S6.5-S6.6). BC emissions in Europe, North America, and Pacific declined by about 30% but their share in the global total ~~are~~ is estimated at below 15% in 2010 (from about 24% in 1990).

### 4.2 Comparison with other studies

This is the first assessment of the global anthropogenic emissions of $PM_{10}$, $PM_{2.5}$, and $PM_1$ using a consistent ~~bottom~~ bottom-up approach across all the sources and regions and therefore only limited comparison to other work at a global level can be made. In fact, the only global set where $PM_{10}$, $PM_{2.5}$, BC~~,~~ and OC were published is the so called 'mosaic inventory'~~,~~ developed within the UNECE Task Force on Hemispheric Transboundary Air Pollution (HTAP) where a compilation of

EDGAR and several regional inventories ~~were~~ was put together (Janssens-Maenhout et al., 2015) for 2010. For most of the species the HTAP_v2 is lower than ECLIPSE *V5a* by about 20-30% except OC where the agreement is good (Table S8.1 in SI). It is difficult to ~~easily conclude~~draw conclusions on the reasons for the observed differences as the methods are not fully comparable and HTAP_v2 is a compilation ~~where~~ in which single products rely on different methods. However, as further discussion shows, the largest discrepancy for $PM_{10}$ and $PM_{2.5}$ is for China, as well as Europe and Russia; the sum of the differences in these three regions represents about 90% and over 50% of all the difference for $PM_{10}$ and $PM_{2.5}$. There have been a number of global studies of BC and OC emissions as well as several regional assessments of $PM_{10}$, $PM_{2.5}$, BC, and OC which we discuss in ~~a~~ more detail below.

A seminal work by Bond et al. (2004) established a benchmark global inventory of BC and OC emissions for the year 1996 that was later updated to 2000 (Bond et al., 2013) and was also used as the basis for the development of BC and OC emissions in the RCP scenarios (Lamarque et al., 2010; Van Vuuren et al., 2011). Bond et al. (2004) provided a thorough review of BC and OC estimates to date and has been used as the primary reference since. We compare our results with Bond et al. (2004, 2013) in Table 9 and Fig. 9 for 1995 and 2000. At a global level, the recent GAINS calculation (*V5a*) shows higher values, which is mostly due to inclusion and re-estimation of a few sources: kerosene wick lamps, gas flaring, use of regional coal statistics for China; Fig. 9 shows the role of these sources in GAINS estimates for 2000, ECLIPSE version totals (see also Fig. S6.1 in SI), and compares them to the range presented in Bond et al. (2013). Even though the global totals fall within the same range, especially when considering the role of newly calculated emissions from kerosene lamps (version *V4a* did not include them), there are often larger differences at a source-sector level, particularly for residential combustion where the largest uncertainties exist in fuel consumption, its allocation between uses and technologies, and emission factors (Table 9). Excluding kerosene lamps and gas flaring, which were not included in Bond et al. (2004, 2013), GAINS global estimates are larger by less than 5% and 15% for 1995 and 2000 than Bond et al. (2004, 2013). This difference is mostly due to the residential sector where comparable source categories are larger in GAINS by 40-60% but the overall balance is partly offset by emissions from industrial coal use (including coke and brick production as well as industrial boilers) that are larger in Bond et al. (2004, 2013) (Table 9).

Emission characteristics for kerosene lamps, gas flaring, and diesel generators have been included in GAINS only recently (most of the previously published global work has not included these sources). For kerosene wick lamps we followed on the work of Lam et al. (2012) but developed an independent assessment of activity data and estimated global BC emissions from this source at 706 Gg in 2005. Our estimates are higher than the previous assessment of 270 Gg (Lam et al., 2012) and 580 Gg (Jacobson et al., 2013) because of larger kerosene consumption in our study, but compare well to Elisabeth (2013) who calculated 702 Gg BC from this activity. For gas flaring we estimated global BC emissions at about 270 Gg and 210 Gg in 2005 and 2010. A recent study of flaring emissions for the Bakken field (Weyant et al., 2016) extrapolated their results to global estimates of 20±6 Gg BC, assuming the same range of emission factors as measured by them at the Bakken field. This is over ten times less than our estimates but we argue that the Bakken flares are not necessarily representative for some of the other regions where strong variability and potentially high soot emissions have been shown by (Conrad and Johnson, 2017;

Johnson et al., 2011) and also speculated in Huang et al. (2015). We found no global estimates of PM emissions from diesel generators and our estimate of 113 Gg for $PM_{2.5}$ and 50 Gg for BC in 2010 confirms that it appears to be a rather small source from a global perspective, and although~~,~~ important locally, it is expected that in the near future with reliable access to grid electricity use of DG sets will be limited particularly in residential, commercial and industrial sectors.

Granier et al. (2011) compared global and regional estimates of BC developed within global and regional modelling activities or inventories for the period 1980-2010. We compare the range presented in that study with the inventory used during development of RCP scenarios (Lamarque et al., 2010) and the GAINS model calculation for version *V5a*, highlighting the role of the newly included and re-estimated sources (Fig. 10). At a global level, the GAINS range overlaps ~~with~~ the span of estimates presented in other studies, although the GAINS total is actually higher than all previous estimates and the post 2000 trend is also different, implying a slight increase in emissions rather than a decline or stabilization shown in earlier studies; note that values reported in Granier et al. (2011) for 2010 were results of projections. As shown in comparison to Bond et al. (2004, 2013) (Table 9), the GAINS values are higher primarily due to inclusion of kerosene lamps and gas flaring but also because of more recent statistical data for 2010 than used in the previously published work. Fig. 10 also includes results of selected global and regional studies which were not explicitly referred to in Granier et al. (2011); these are marked with 'black star' symbols and included in Table S8.1 in SI. The values for 1996 and 2000 refer to ~~the~~ Bond et al. (2004, 2013) and for 2010 to the HTAP_v2 inventory (Janssens-Maenhout et al., 2015), none of which included emissions from kerosene wick lamps.

Fig. 10 shows also a similar comparison for selected countries: China, India, and US; note that the ranges presented in Granier et al. (2011) for regions/countries do not necessarily add up to the global total as the former included also selected regional studies which were not part of the comparison of the global totals. For China, a continuing growth in BC emissions has been reported in all investigated studies. GAINS is comparable with the RCP input (Lamarque et al., 2010) for 1990-1995, while for the last decade it is consistently higher or at the top of the range, which in Granier et al. (2011) is representative of the upper estimates in the RCP scenarios rather than specific inventories. However, a number of recently published studies for China reported rather high BC, e.g., Zhang et al. (2009) estimated about 1.8 Tg for 2006, HTAP_v2 (based on the MEIC[17] system developed by the Tsinghua University (Beijing, China)) 1.76 Tg for 2010, Lu et al. (2011) 1.84 Tg for 2010, and 1.92 Tg for 2008 using a top-down approach (Kondo et al., 2011); these results and other recent regional studies are marked with 'black star' symbols in Fig. 10 and included in Table S8.1. Several authors estimated ~~also~~ $PM_{10}$ and $PM_{2.5}$ emissions for China and these compare reasonably well with GAINS, although they are systematically lower by up to 15% with the exception of the HTAP_v2 mosaic inventory (Janssens-Maenhout et al., 2015) which is lower by nearly 25% for 2010 (Table S8.1 in SI); the latter inventory relies on the data from the MEIC system where more optimistic assumptions about the penetration and achieved efficiency of wet scrubbers and electrostatic precipitators in industry are made. For India, all inventories suggest emissions have been increasing in the investigated period but there is a very large

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[17] MEIC - Multi-resolution Emission Inventory for China; http://www.meicmodel.org

spread of estimates. Current GAINS estimates are higher than Lamarque et al. (2010) and the range shown by Granier et al. (2011) (Fig. 10) – the overlap in the last decade is because the upper values are based on the earlier GAINS model estimates (e.g., Klimont et al., 2009) which are consistent with ECLIPSE set. Some recent papers have shown similar BC emissions ~~as~~ to GAINS (e.g., Janssens-Maenhout et al., 2015; Lu et al., 2011; see also Table S8.1) but overall the range of published emission estimates for PM species for India varies greatly between studies, e.g., for BC from about 350 Gg to over 1000 Gg (Table S81). A lot of that variability links to different assumptions about biomass use for cooking (Venkataraman et al., 2005), efficiency of PM abatement in power and industry, and large uncertainty in agricultural burning activity (Venkataraman et al., 2006). For the US, all studies indicate a declining trend in BC emissions (Fig. 10). However, in ~~contrary~~ contrast to China and India, GAINS emissions are in the lower range of existing estimates (Fig. 10, Table S8.1) and differences in emissions from non-road machinery and agricultural (or prescribed) burning appear~~s~~ to be the key reason for observed discrepancies.

For Europe (including European part of Russia), the published studies of BC and OC (Bond et al., 2004; Kupiainen and Klimont, 2007; Schaap et al., 2004; see Table S8.1) compare well showing differences within ±10% or less with the exception of EDGAR (Janssens-Maenhout et al., 2015) which shows much lower emissions but does not include any Russian territory. At the level of whole of Europe, GAINS calculates similar PM$_{10}$ and PM$_{2.5}$ emissions as officially reported to UNECE LRTAP Convention (www.ceip.at), while the EDGAR estimate is nearly 40% lower for both species~~,~~ ~~however,~~but does not include Russia (Table S8.1). There have been only few published estimates of PM emissions in Russia (Table S8.1). For PM$_{10}$ and PM$_{2.5}$ in 2010, GAINS calculates higher emissions than EDGAR (Janssens-Maenhout et al., 2015) or the national inventory submitted to LRTAP Convention (www.ceip.at) which covers only the European part of Russian Federation; remarkably, the total EDGAR estimate is similar to the national submission for the European part. The main reasons for discrepancy are significantly larger GAINS emissions from industrial processes, residential combustion (these are very low in the national submission – less than a quarter of EDGAR and GAINS estimates), agricultural burning, as well as inclusion of gas flaring. The uncertainties in volume of gas flared and actual emission factors are major reasons for the difference in estimated BC emissions in GAINS and Huang et al. (2015), who derived a much higher emission factor for this activity; for other sectors both studies report ~~a~~ fairly similar emissions of BC for 2010.

Yan et al. (2011) developed projections of PM$_{10}$ emissions from the road transport sector (exhaust only). Their PM$_{10}$ estimates for 2000-2010 were about 1.65-1.75 Tg with a contribution from high emitters of about 0.3 Tg. The ECLISPE *V4a* results are comparable to Yan et al. (2011), while in *V5* and *V5a*, updates to the emission factors (reflecting more recent measurements, poor fuel quality, and maintenance) and penetration rates of control measures for developing countries (often delayed or postponed implementation of legislation) led to higher estimates of about 2.4-2.6 Tg, including high emitters (0.4-0.5 Tg). Total GAINS model estimates for road transport also include non-exhaust emissions (brake, tyre, road abrasion) which add up to around 0.6 Tg PM$_{10}$.

Wiedinmyer et al. (2014) developed a new assessment of global emissions from burning of waste, including particulate matter. That study suggests that all current estimates largely underestimate emissions from this activity. Compared to

GAINS, their emissions are nearly seven times larger and would make open burning of waste one of the key categories contributing between 10-15% of BC and $PM_{2.5}$ and nearly 30% of OC~~;~~ considering anthropogenic sources. For example, waste burning could be responsible for three times more emissions of BC, OC, and $PM_{2.5}$ than agricultural waste burning or about a third of the total transport sector emissions. Current GAINS estimates of 2010 emissions from open waste burning are about 1.4, 1.3, 0.1, 0.75 Tg $PM_{10}$, $PM_{2.5}$, BC, OC, while Wiedinmyer et al. (2014) calculated for the same species 12, 12, 0.632, 5.1 Tg. Obviously, large uncertainties remain in activity data and actual emission factors (see discussion in section 3.8) but this activity deserves more attention in the future.

### 4.3 Uncertainty in emission estimates

The completeness and quality of information about emission inventories varies across the regions, sectors~~,~~ and species. The underlying information about several key PM sources like residential solid fuel combustion, brick production, and residual waste burning is often of poor quality or non-existing and that applies to both activity data and emission factors. In order to create a comprehensive emission data set, the national information is often supplemented with model estimates that rely on default parameterization; in fact, even many of the national inventories draw on the international data sets of emission factors (e.g., EEA, 2013; US EPA, 1995) owing to lack of local measurements. Finally, the level of enforcement of existing laws, as well as the real-life performance of control technology is seldom sufficiently well-known and we tend to assume rather optimistically that both deliver and work as planned which has been shown to be often false (e.g., Stoerk, 2016; Xu et al., 2009; Xu, 2011) as, more recently, in the so-called Dieselgate affair (e.g., Lange and Domke, 2015; US EPA, 2015a, 2015b). Consequently, the level of uncertainty, or confidence, varies widely across source sectors and regions.

We have not performed a formal uncertainty analysis for emission estimates in this study, but results of analysis from other studies are helpful and indicative of the expected uncertainties for various species and regions. For example, the global BC and OC inventory developed by Bond et al. (2004) included an uncertainty analysis of total emissions providing regional 'low-high' estimates for 1996. For BC emissions from anthropogenic sources, the range was 3.1-10 Tg $yr^{-1}$ (-30% to +120%) and for OC 5.1-14 Tg $yr^{-1}$ (-40% to +130%). Estimates from the GAINS model presented in this study sit well within these ranges.

As indicated earlier, emissions of PM, including carbonaceous aerosols, belong to the most uncertain among ~~the~~ air pollutants, as they form usually under poor combustion conditions in small inefficient installations burning poor quality fuels, which brings variability to the emission characteristics. Additionally, there is very little information globally about local emission factors. Considering local data and knowledge about emission sources and their emission factors could significantly reduce uncertainties (Zhang et al., 2009). Allocating total PM emissions into different size bins or chemical species (here BC and OC) is associated with uncertainties that for a specific source are determined by the measurement. Among others, Bond et al. (2013) discussed specific issues related to BC and OC aerosols, while for PM size distribution there exists specific analysis for particular measurement equipment (e.g., Armas et al., 2007; Coquelin et al., 2013) and most of the studies reporting measurements of size distribution estimate uncertainties for each size category. While the sum of all the PM species is constrained by the total mass, the single size distribution values rely on a large number of measurements reducing the overall uncertainty. Exceptions are source~~-~~ sectors for which very few measurements exist, e.g., coke ovens, fireworks, handling of bulk materials.

In addition to the emission characteristics, the activity data are also~~ is~~ a source of uncertainty. While for major industrial and transport sectors there are ~~well ~~well-documented and regularly updated national and international sources of activity data (e.g., IEA, 2015a, 2015b), the activities behind the major PM source categories, for example poor quality fuels in cook stoves or brick kilns, as well as local vehicle fleets, are not well known. For commercial fuels, however, the uncertainty has been estimated to vary from 2-3% for OECD countries to 5-10% for non-OECD (IPCC, 2006a).

A significant part of total aerosol emissions originate from open biomass burning, including forest fires, savannah, and agricultural residue burning (e.g., Reddington et al., 2015). Estimation of activity data and actual emission factors are bound with significant uncertainties which include, among others, amount of biomass burned and interannual variability (Chen et al., 2013; van der Werf et al., 2006; Wiedinmyer et al., 2011), drivers and impact of change in agricultural fires (Morton et al., 2008), and emission factors (Castellanos et al., 2014). The uncertainty ranges estimated by Bond et al. (2004) for BC and OC emissions from open biomass burning were 1.6 to 9.8 Tg yr$^{-1}$ (-45% to +185%) for BC and 31 to 58 Tg yr$^{-1}$ (-40% to +110%) for OC.

The uncertainties of emission estimates developed with integrated assessment models like GAINS are similar to the estimates for bottom-up inventories discussed above, at least at a regional scale. Additionally, ~~the ~~error compensation, which is especially relevant if calculated emissions are the sum of a large number of equally important source categories (and where the errors in input parameters are not correlated with each other), can lead to a further reduction of overall emission uncertainty (Schöpp et al., 2005). A careful assessment of the assumption about correlation between input parameters is essential as, for example, poor enforcement of legislation or measurement~~s~~ errors could affect several source sectors in a similar way. The GAINS model uncertainties, calculated in Schöpp et al. (2005), are consistent with the values reported by Streets et al. (2003) for developed countries. This analysis has also shown that at a finer scale the understanding of local circumstances are critically important to reduce uncertainty, and while the emission factors were estimated to be the key factor determining uncertainty in historical emissions, at least for aerosol emissions, the uncertainty in activity assumptions becomes more important for the uncertainties in projected emissions.

## 5 Conclusions

To our knowledge, the estimates represent the first global dataset of anthropogenic emissions where ~~size ~~size-specific mass PM calculation, including BC and OC, was performed using a uniform and consistent estimation framework including a number of previously unaccounted or often misallocated emission sources, i.e., kerosene lamps, gas flaring, diesel generators~~,~~ and trash burning that have been systematically evaluated for each region. Spatially, emissions were calculated for 170 regions and allocated to $0.5^{o}$ x $0.5^{o}$ longitude-latitude grids and are available either from the on-line GAINS model[18], where assumptions and results can be displayed for 25 global regions (see section S7 in SI) or gridded emissions can be downloaded from the project website[19]. The ECLIPSE datasets do not include independent estimates of emissions from forest fires and savannah burning, windblown dust, and unpaved roads.

We estimate that global emissions of PM have not changed much between 1990 and 2010 but there are significantly different regional trends with North America, Pacific, and Europe reducing emissions by 30 to over 50%, and Asia and Africa increasing by about 30%. While these regionally varying developments are clearly visible in $PM_{2.5}$ and $PM_{10}$ estimates, the BC regional changes were somewhat less dramatic, mostly because trends in power and industrial sector emissions of PM are much less relevant for total black carbon emissions. Globally, over 75% of anthropogenic $PM_{10}$ and $PM_{2.5}$ originates from residential combustion, power plants and industry, while for BC residential combustion and transport represent more than 75% but the importance varies across regions with Europe and North America having transport as key, and the rest of the world residential combustion. Our new global estimate of BC emissions suggests higher numbers than previously published owing primarily to inclusion of new sources.

We argue that this PM estimate reduces the gap in source coverage required in air quality and climate modelling studies and health impact assessments at a regional and global level as it includes both carbonaceous and non-carbonaceous constituents of primary particulate matter emissions~~, ~~; however, additional efforts need to be made to address several fugitive sources of anthropogenic dust, e.g., unpaved roads. The ECLIPSE emission data sets have been used in several regional and global atmospheric transport and climate model simulations (AMAP, 2015; Eckhardt et al., 2015; Gadhavi et al., 2015; Lund et al., 2014; Quennehen et al., 2016; Stohl et al., 2013, 2015; Wobus et al., 2016; Yttri et al., 2014) where various aspects of several particulate matter species were addressed. The emissions developed during ECLIPSE also served as the basis for a recently published global particulate number estimate~~s~~ (Paasonen et al., 2016).

We envisage development of further datasets drawing on the experience of the ECLIPSE exercise. The future versions will be available via the same on-line platform where additional documentation will be placed too. As a matter of fact, the GAINS model and the ECLIPSE dataset and scenarios have already been ~~already~~ used as a starting point to develop emission data and mitigation strategies for the recently published International Energy Agency (IEA) World Energy Outlook special report on air pollution (IEA, 2016). Furthermore, ~~the~~ elements of the ECLIPSE data have been part of the contribution towards improved representation of carbonaceous aerosols in the ~~large~~ large-scale integrated assessment models used in the development of the Shared Socio-economic Pathways (SSP) (O'Neill et al., 2014; Rao et al., 2017; Riahi et al., 2017).

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[18] http://magcat.iiasa.ac.at/gains/IAM/index.login

[19] http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html

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**Table 1.** Overview of the ECLIPSE emission data sets available to date

| **Version** | **Release date** | **Period covered** | **Comments; key features** |
|---|---|---|---|
| V3 | Nov 2013 | 2005, 2008, 2009, 2010 | Estimates for 2008 and 2009 based on activity proxies and trends in internationally reported emissions; activity data for 2010 based on the IEA World Energy Outlook 2011 (IEA, 2011) |
| V4a | Jan 2014 | 2005, 2010, *2030*, *2050* | Major updates of EU-28 data (Amann et al., 2012) |
| V5 | Apr 2014 | 1990-2010[a], *2015-30*[a], *2040*, *2050* | IEA and FAO statistical data reimported for the period 1990-2010, international shipping included |
| V5a | Jul 2015 | 1990-2010[a], *2015-30*[a], *2040*, *2050* | China 12th 5-year plan included, improved regional resolution for Latin America, update of: global cement legislation, gas flaring, OC/OM ratios for residential combustion in Asia, Africa, Latin America, EU-28 update (Amann et al., 2015) |

[a] Estimated in 5-year intervals

**Table 2.** Residential-commercial sector fuel and source structure in GAINS

| | | | | | | Household boiler | | Medium boiler | |
|---|---|---|---|---|---|---|---|---|---|
| Fuels | Non-specific | Lighting | Three-stone | Fireplace | Stove [a] | Manual | Auto | Manual | Auto |
| Gaseous fuels | ● | | | | | | | | |
| Liquid fuels | ● | ● | | | | | | | |
| Charcoal | ● | | | | | | | | |
| Coal | | | | | ● | ● | ● | ● | ● |
| Biomass | | | | | | | | | |
| - Fuelwood | | | ● | ● | ● | ● | ● | ● | ● |
| - Agricultural residue | | | ● | | ● | | ● | | ● |
| - Dung cake | | | ● | | ● | | | | |

[a] distinguishing cooking and heating stoves as separate categories

**Table 3.** Mitigation measures distinguished in the residential-commercial sector in GAINS

| | | | | | Stove | | Household boiler | | Medium boiler | |
|---|---|---|---|---|---|---|---|---|---|---|
| Control option | Non-specific | Lighting | Three-stone | Fireplace | Cooking | Heating | Manual | Auto | Manual | Auto |
| Improved | • | | | • | • | • | • | | | |
| New | | | | • | • | • | • | | | |
| Fan stove | | | | | • | | | | | |
| Coal briquettes | | | | | • | • | | | | |
| Hurricane lamp | | • | | | | | | | | |
| LED [a] lamp | | • | | | | | | | | |
| Pellets | | | | | | • | • | • | • | • |
| Cyclone | | | | | | | | | • | • |
| ESP [b] | | | | | | • | • | • | | • |

[a] Light Emitting Diode

[b] Electrostatic precipitator

**Table 4.** Brick sector technology structure assumed in GAINS for different regions

| Kiln type | East Asia [a] | South-East Asia [b] | Central Asia | Africa | Latin America and Caribbean | Other |
|---|---|---|---|---|---|---|
| Traditional clamp | • | • | • | • | • | |
| Downdraft | | • | • | • | • | |
| Moving chimney Bull's trench | | • | | | | |
| Fixed chimney Bull's trench | | • | | | | |
| Zig-Zag | | • | | • | | |
| Vertical shaft brick kiln | • | • | | • | • | |
| Marques kiln | | | | | • | |
| Hoffman kiln | • | • | • | • | • | |
| Tunnel kiln (coal) | • | • | • | • | • | • |
| Tunnel kiln (gas, oil) | • | • | • | • | • | • |

[a] Excluding OECD countries which are included in 'Other'

[b] Including Middle East

**Table 5.** Overview of sectoral layers included in the gridded ECLIPSE emissions of PM

| **Sector layer** | **Included activities** |
|---|---|
| Energy [a] | Power plants, energy production/conversion, fossil fuel distribution |
| Industry | Industrial combustion and processes |
| Residential | Residential and commercial combustion sources |
| Transport [b] | Road and non-road transport sources; including tyre and brake wear, road abrasion |
| Waste | Waste disposal, including trash burning |
| Agriculture | Livestock and arable land operations (ploughing, harvesting) |
| Agriculture (open burning) [c] | Open burning of agricultural residues (excluding forest and savannah burning) |
| Total | The sum of the above sectors |
| Shipping [d] | International shipping; available in version *V5* and *V5a* |

[a] Includes associated petroleum gas flaring which is also available as a separate gridded layer

[b] Does not include resuspension and international air and shipping; for the latter recommendation to use the RCP datasets, except for version *V5* and *V5a* where international shipping was also included

[c] The gridding proxy has been acquired from the GFED3.1 (van der Werf et al., 2010)

[d] Available as a separate file where all pollutants' emissions are included; the resolution of this layer is $1^{o}$x$1^{o}$

**Table 6.** Particulate matter amplification factors for high emitting light- and heavy-duty diesel and gasoline vehicles used in the GAINS model

| | Light duty | | Heavy duty | |
|---|---|---|---|---|
| | diesel | gasoline | diesel | gasoline |
| No control | 3 | 6 | 3 | 4 |
| Euro 1/I | 3 | 6 | 3 | 4 |
| Euro 2/II | 5 | 6 | 5 | 10 |
| Euro 3/III | 5 | 10 | 5 | 10 |
| Euro 4/IV | 5 | 10 | 5 | - |
| Euro 5/V | 10 | 10 | 10 | - |
| Euro 6/VI | 10 | 10 | 10 | - |

**Table 7.** Regional emissions of particulate matter in 2010, ECLIPSE *V5a*, Gg year$^{-1}$

| | $PM_{10}$ | $PM_{2.5}$ | $PM_1$ | BC | OC | OM |
|---|---|---|---|---|---|---|
| Africa | 9161 | 7973 | 6959 | 1347 | 3023 | 5207 |
| East Asia | 27172 | 20241 | 15291 | 2622 | 4974 | 7996 |
| Europe and Russia | 6027 | 4105 | 2781 | 660 | 897 | 1399 |
| Latin and Central America | 3736 | 2947 | 2358 | 508 | 994 | 1617 |
| North America | 1964 | 1268 | 917 | 249 | 382 | 594 |
| Pacific | 609 | 347 | 220 | 62 | 75 | 115 |
| South-West and Central Asia | 11982 | 9174 | 7654 | 1686 | 2796 | 4667 |
| International shipping | 1856 | 1758 | 1612 | 120 | 398 | 517 |
| International aviation [a] | 30 | 30 | 28 | 10 | 10 | 13 |
| Global anthropogenic | 62537 | 47843 | 37819 | 7264 | 13548 | 22125 |
| Forest and savannah fires [b] | 48207 | 33014 | 33014 | 2268 | 19489 | 31363 |
| Global total | 110744 | 80858 | 70834 | 9532 | 33037 | 53489 |

[a] Values are middle of the range estimates referring to the ranges reported in Settler et al. (2013), Yim et al. (2015), and based on global fuel consumption and ranges of emission factors from Kinsey (2009)

[b] GFED3.1 without agricultural waste burning; $PM_{10}$ value based on TPM (total particulate matter); $PM_1$ not available in GFED – here assumed equal $PM_{2.5}$

**Table 8.** Sectoral emissions of particulate matter in 2010, ECLIPSE *V5a*, Gg year$^{-1}$

| | $PM_{10}$ | $PM_{2.5}$ | $PM_1$ | BC | OC | OM |
|---|---|---|---|---|---|---|
| Agriculture | 6555 | 3848 | 2883 | 337 | 1313 | 2364 |
| Residential combustion | 23078 | 21857 | 20742 | 4163 | 8852 | 15329 |
| Industrial processes | 12162 | 8340 | 4135 | 462 | 633 | 823 |
| Large scale combustion | 11561 | 6420 | 3812 | 136 | 164 | 248 |
| Oil & gas, mining | 1706 | 571 | 412 | 226 | 93 | 120 |
| Transport – road | 3339 | 2925 | 2524 | 1349 | 1116 | 1451 |
| Transport – non-road | 861 | 823 | 795 | 363 | 217 | 283 |
| Waste | 1388 | 1272 | 876 | 97 | 751 | 977 |
| International shipping | 1856 | 1758 | 1612 | 120 | 398 | 517 |
| International aviation [a] | 30 | 30 | 28 | 10 | 10 | 13 |
| Global anthropogenic | 62537 | 47843 | 37819 | 7264 | 13548 | 22125 |
| Forest and savannah fires [b] | 48207 | 33014 | 33014 | 2268 | 19489 | 31363 |
| Global total | 110744 | 80858 | 70834 | 9532 | 33037 | 53489 |

[a] Values are middle of the range estimates based on the ranges reported in Settler et al. (2013), Yim et al. (2015), and based on global fuel consumption and ranges of emission factors from Kinsey (2009)

[b] GFED3.1 without agricultural waste burning that is included based on GAINS estimates in category 'Agriculture'; $PM_{10}$ value based on TPM (total particulate matter); $PM_1$ not available in GFED – here assumed equal $PM_{2.5}$

5

**Table 9.** Comparison of global anthropogenic emissions of BC by sector, Gg year$^{-1}$

| | 1995 | | 2000 | |
|---|---|---|---|---|
| | Bond et al. (2004)[a] | This study (*V5a*) | Bond et al. (2013) | This study (*V5a*) |
| Diesel engines – road | 792 | 872 | 840 | 980 |
| Diesel engines – off-road | 579 | 415 | 470 | 432 |
| Residential combustion | 2046 | 3703 | 1880 | 3891 |
| *of which:* | | | | |
| *Biomass cooking* | *1481* | *1660* | *1290* | *1711* |
| *Biomass heating* | | *411* | *260* | *392* |
| *Residential coal* | *480* | *710* | *330* | *908* |
| *Other* [b] | *85* | *922* | [c] | *880* |
| Agricultural burning | 328 | 323 | 330 | 326 |
| Industrial coal [d] | 642 | 282 | 740 | 315 |
| Other [e] | 610 | 612 | 600 | 649 |
| Global anthropogenic | 4997 | 6206 | 4870 | 6594 |

[a] Estimates for 1996

[b] GAINS includes oil appliances and kerosene lamps – the latter are estimated in GAINS at 750 and 692 Gg BC in 1995 and 2000

[c] Other residential sources (oil) included in category 'Other'

[d] Includes coke and brick production, coal boilers and furnaces

[e] Includes power plants, gas flaring, waste, gasoline engines in transport; for Bond et al. also oil use in residential sector

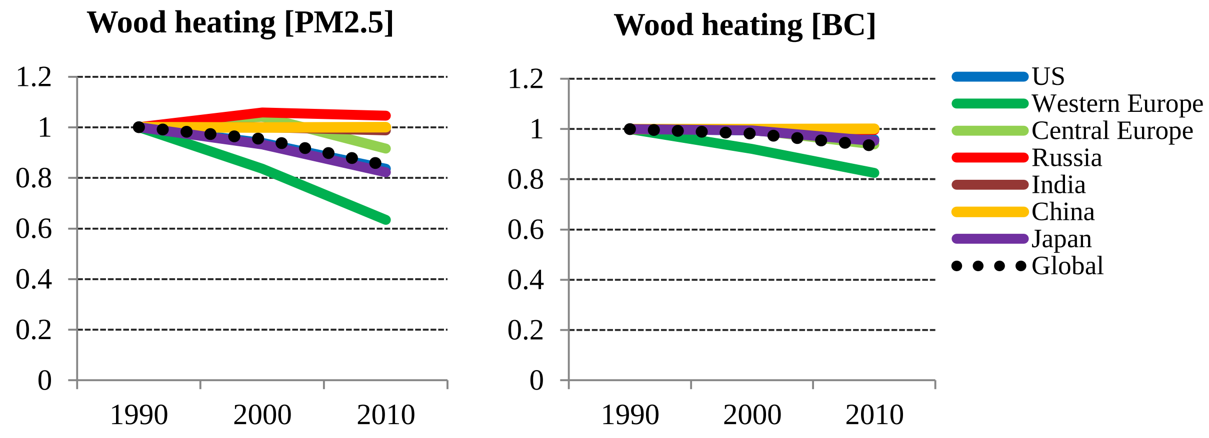


**Figure 1.** Change in implied $PM_{2.5}$ and BC emission factors for residential wood heating in selected countries and world regions; changes relative to 1990 in ECLIPSE *V5a* dataset.

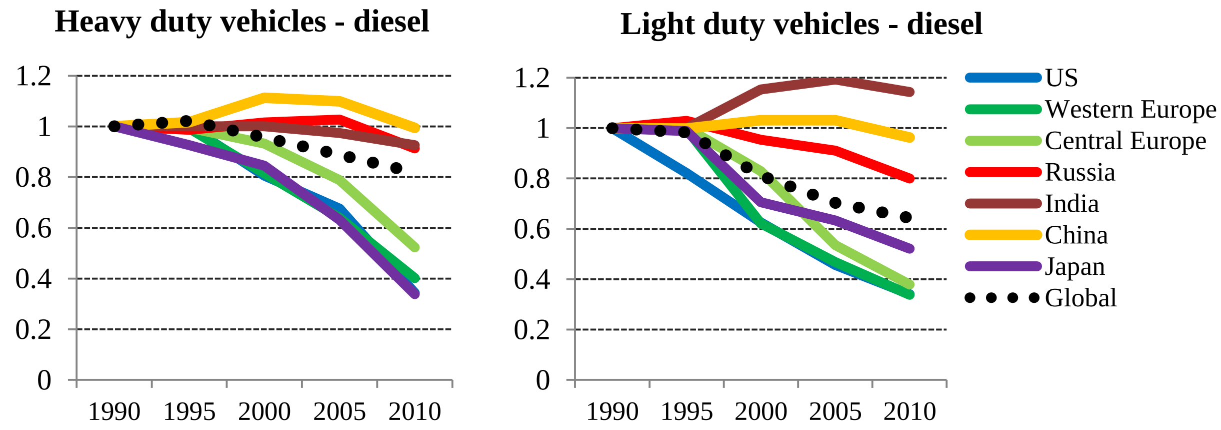


**Figure 2.** Change in implied BC emission factors for road diesel vehicles in selected countries and world regions; changes relative to 1990 in ECLIPSE *V5a* dataset.

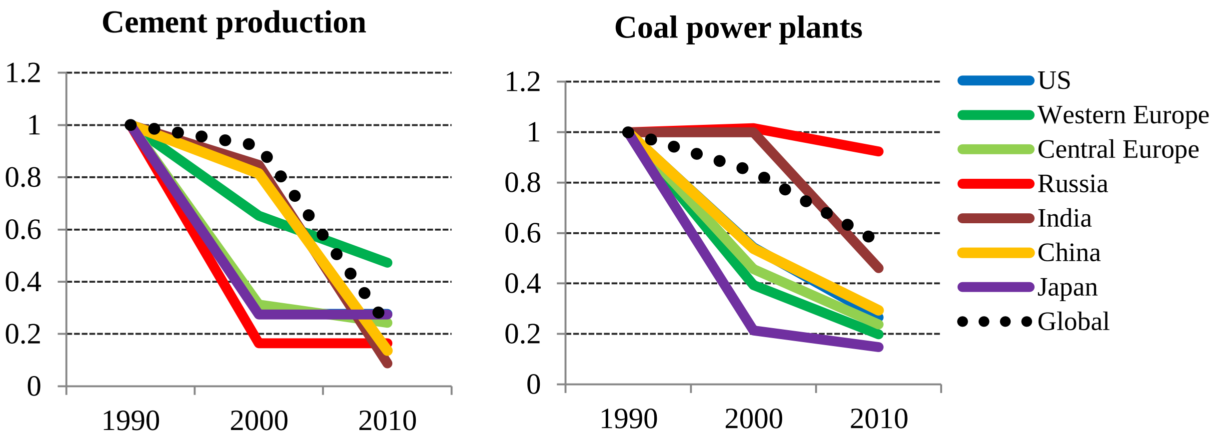


**Figure 3.** Change in implied $PM_{2.5}$ emission factors for cement production and coal power plants in selected countries and world regions; changes relative to 1990 in ECLIPSE *V5a* dataset.

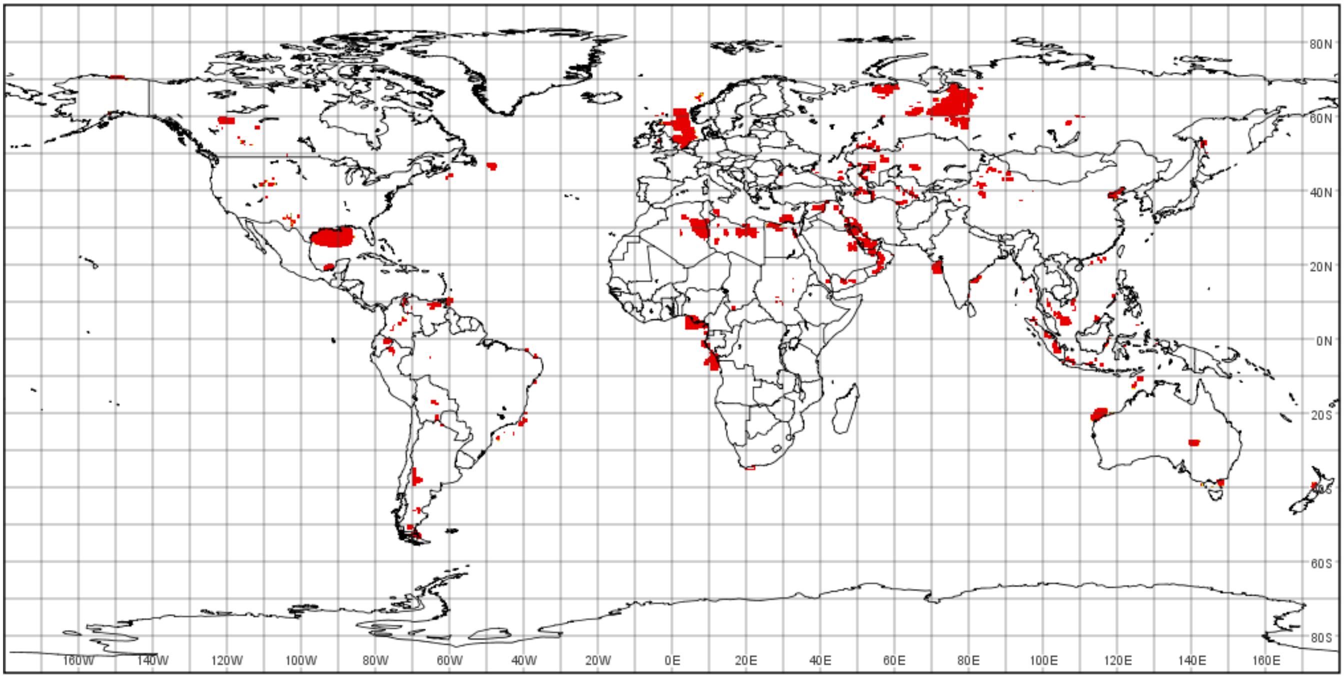
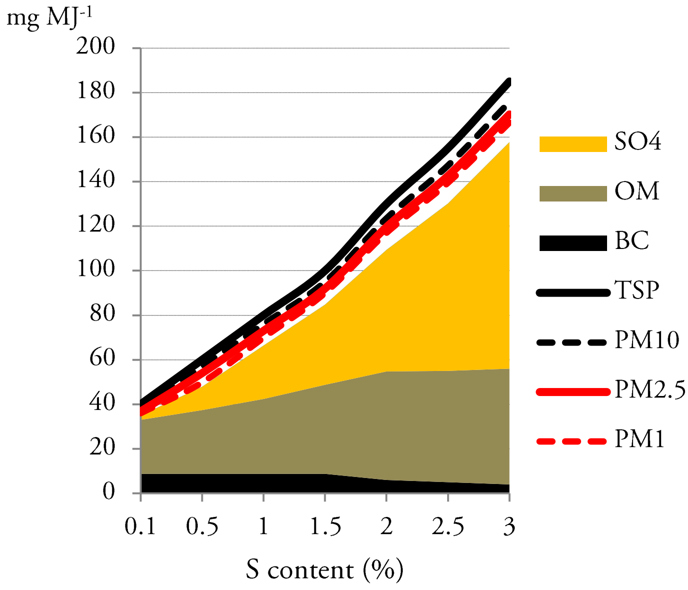


**Figure 4.** Global distribution of grids (0.5°x0.5°) for which flaring of associated petroleum gas emissions were calculated; derived from the 2009 data from Elvidge et al. (2011).



PM emission factors
(mg MJ$^{-1}$)

| | Sulphur content (%S) | | | | |
|---|---|---|---|---|---|
| | 0.1 | 0.5 | 1 | 2 | 3 |
| $PM_{10}$ | 38 | 57 | 76 | 124 | 176 |
| $PM_{2.5}$ | 37 | 54 | 73 | 120 | 167 |
| BC | 8.8 | 8.8 | 8.8 | 6 | 4 |
| OC | 17 | 20 | 24 | 35 | 37 |

**Figure 5.** Particulate matter emission factors for shipping used in the GAINS model

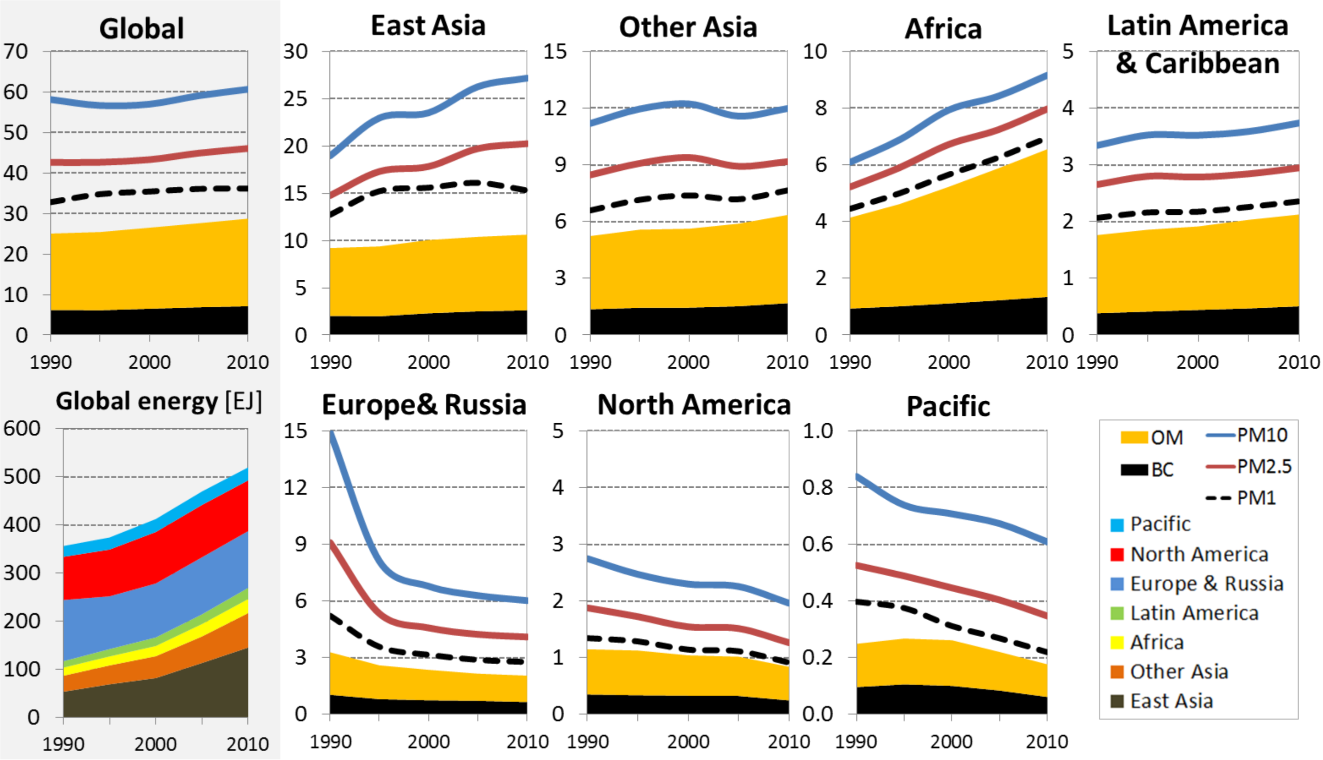


**Figure 6.** Global and regional emissions of PM species [Tg] and global energy consumption [EJ] in the period 1990-2010, ECLIPSE *V5a*.

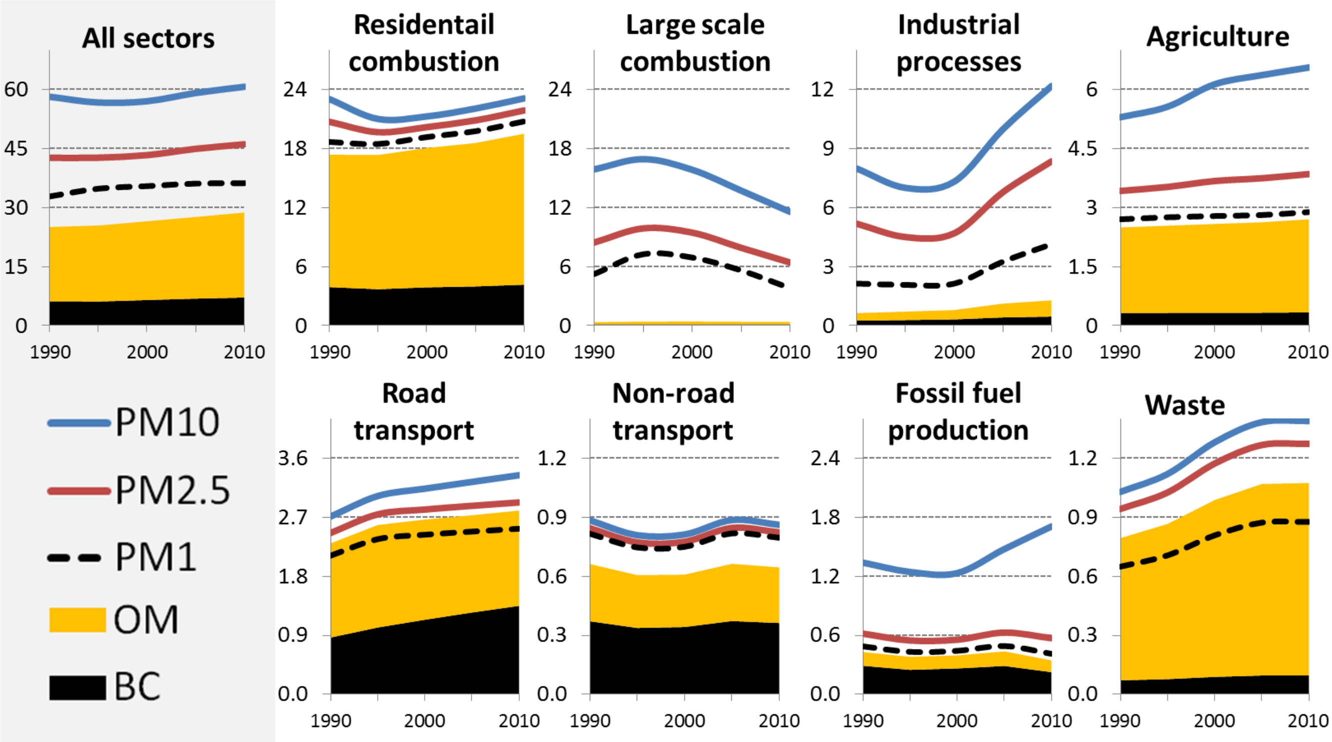
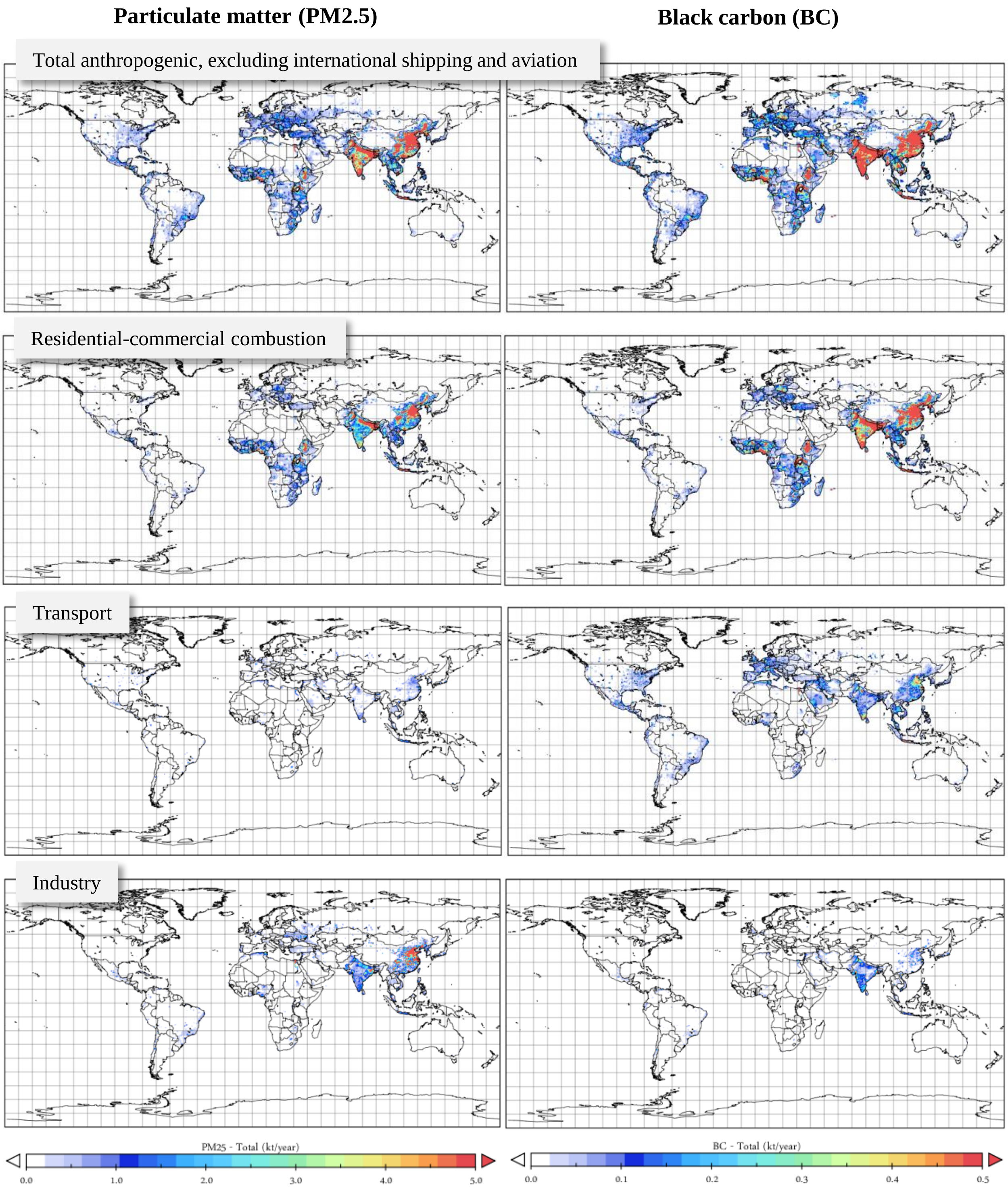


**Figure 7.** Global and sectoral emissions of PM species [Tg] in the period 1990-2010, ECLIPSE *V5a*.



**Particulate matter (PM2.5)**

**Black carbon (BC)**

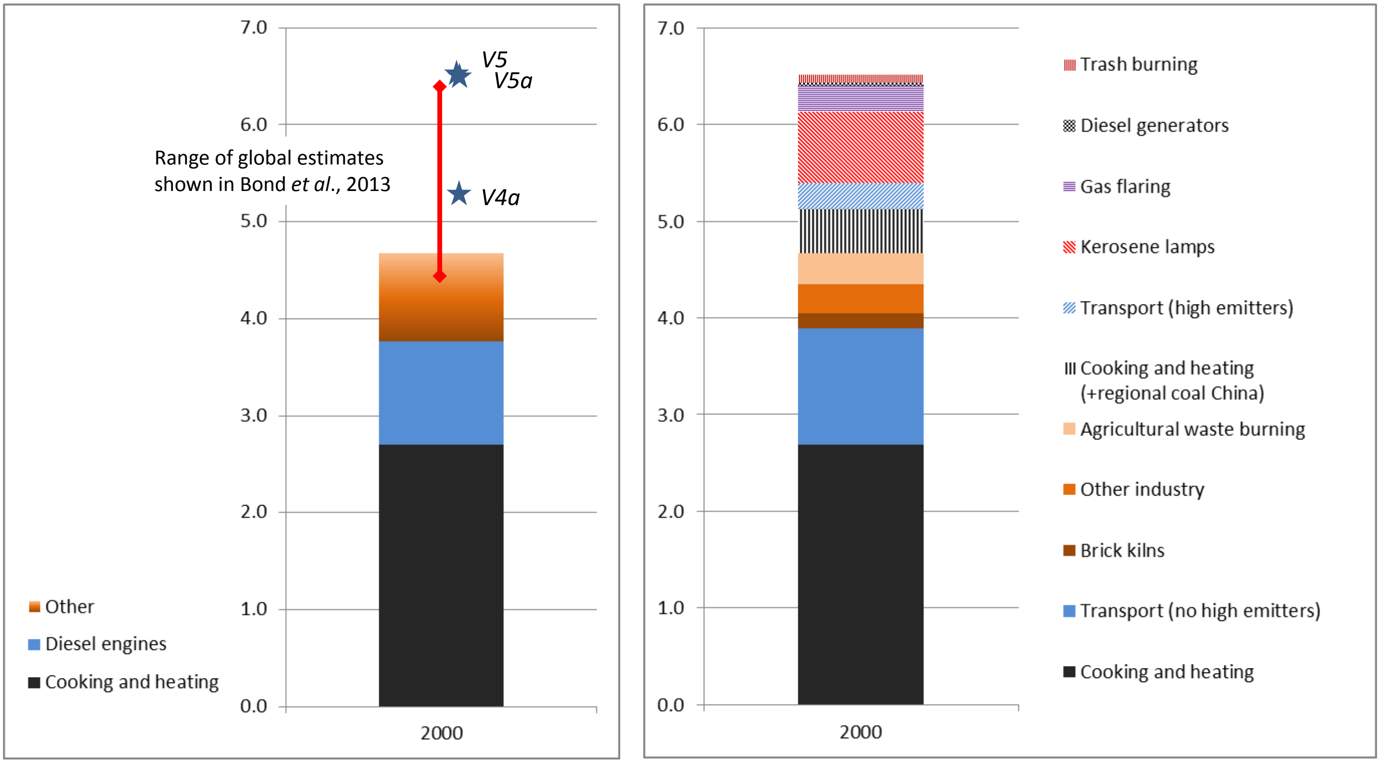
Total anthropogenic, excluding international shipping and aviation

Residential-commercial combustion

Transport

Industry

**Figure 8.** Global distribution of emissions of $PM_{2.5}$ (left) and BC (right) in 2010 [Gg $year^{-1}$ per grid] from land-based sources, ECLIPSE *V5a*; the scale is the same across sectors but there is a factor ten between $PM_{2.5}$ and BC



GAINS; <u>excluding</u> 'new/re-estimated' sources

GAINS; all sources

**Figure 9.** Source-sector distribution of global anthropogenic emission of BC estimated with the GAINS model (ECLIPSE V5a) for the year 2000, Tg year$^{-1}$

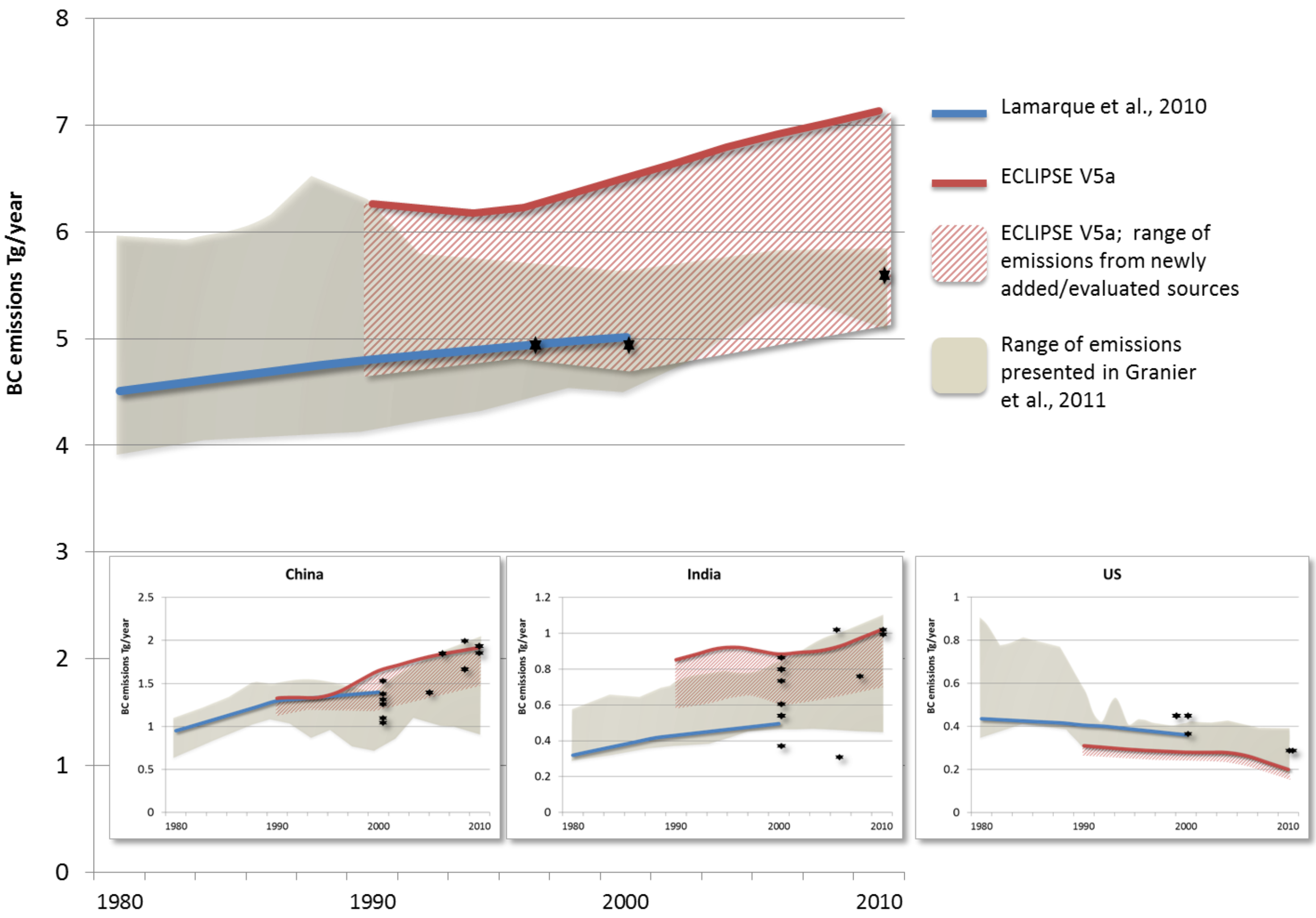


**Figure 10.** Comparison of black carbon emission in this work (ECLIPSE *V5a*) with Lamarque et al. (2010) and Granier et al. (2011). The black star (✸) symbols show emissions reported in global and regional studies listed in Table 8.1 in the SI.

*Supplement of*

# Global anthropogenic emissions of particulate matter including black carbon

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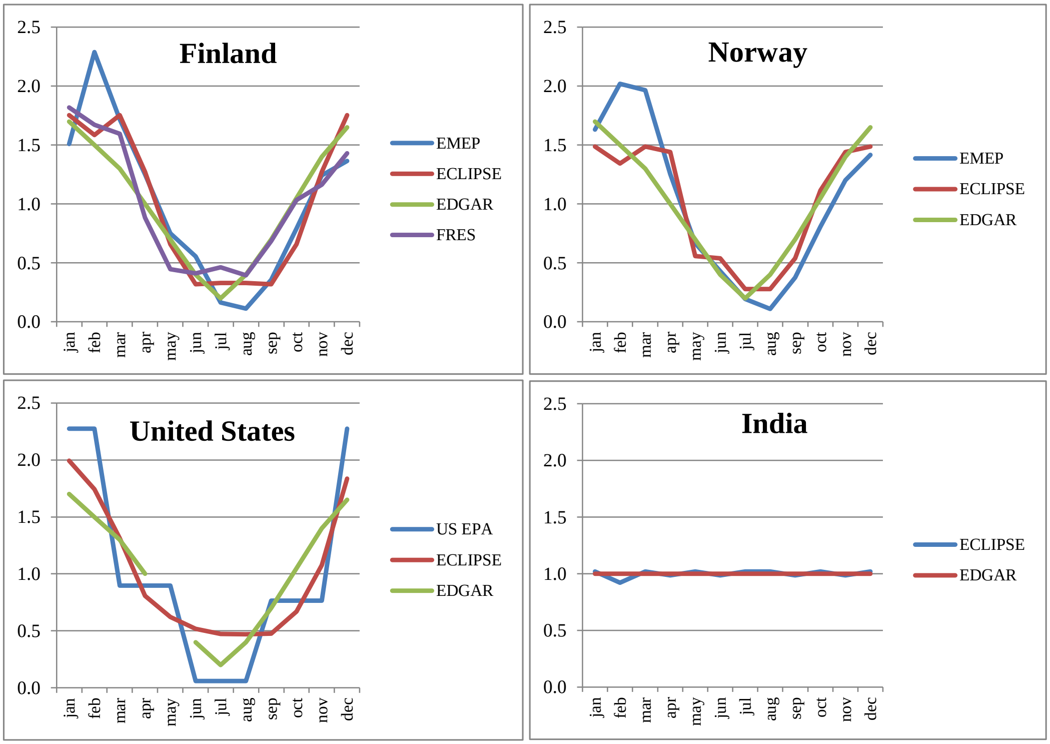
## S1 Comparison of temporal distribution patterns

Fig S1.1 shows a comparison of the temporal patterns (it is an aggregate as the actual patterns are grid specific) for residential combustion sector, applied in the ECLIPSE project, with other data for selected countries.



Finland

Norway

United States

India

**Figure S1.1.** Comparison of monthly distribution of emissions used in ECLIPSE with profiles from EDGAR (EC-JRC/PBL, 2010), EMEP (http://emep.int/mscw/), national Finish model FRES (Karvosenoja, 2008), and US EPA**.**

## S2 Particulate matter emission factors for residential combustion

The GAINS model distinguishes three principal solid fuel stove categories: *traditional*, *improved* and *new stoves*. *Traditional heating stoves* using wood or coal as fuel have simple grate based firebox designs with usually only primary air supply and no heat storing components. Consequently there is restricted availability of air for combustion and poor mixing of air and pyrolysis gases. *Traditional stoves* in general have very high PM emission factors compared with more advanced technologies, but within this category the variability in the emission factors is also large. For example highest emission factors for traditional wood stoves have been measured in situations with restricted combustion air supply that leads to lower burn rate (Jordan and Seen, 2005). Such conditions might prevail when the user wants a lower heat supply to the room. *Improved stoves* have secondary air supply and heat storing components in the firebox construction that improve the combustion performance and reduce emissions of PM compared with the *traditional stoves*. *New stoves* represent the most advanced stove models on the market that have firebox, construction and airflow characteristics that optimize combustion efficiency. Additionally, an electrostatic precipitator (ESP) can be fitted into the latest stoves, which further improve the PM emission performance. GAINS distinguishes also *wood pellet stoves*. Pellets are a very homogenous fuel and combustion is more optimized than batch fired wood log stoves and thus also the PM emissions are lower than with wood log stoves.

A stove heats the surrounding room, but a boiler heats water to be circulated through a piping system to heat an entire house (Johansson et al., 2004). In *old-type wood log boilers* up-draught combustion is commonly used, which resembles the combustion in a stove; *modern wood boilers*, however, use downdraught combustion and often have an isolated burn-out zone (Johansson et al., 2004). In contrast to stoves, wood boilers can be connected to a water tank to store heat, which allows the boiler to be run at a regular heat output and to certain extent optimizing the combustion conditions. Storage tanks are common in modern wood boilers and also old boilers may be equipped with them, leading to lower emissions and higher efficiencies (Johansson et al., 2004). The single family house boilers are typically smaller than 50 $kW_{th}$, the larger residential boilers are allocated to a category *medium size boilers* where manual and automatic boilers are distinguished (Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007). Such boilers might be an important emission source, especially when many of them are fired with coal, but there are not a of lot measurements available. The GAINS model relies on studies discussed previously (EEA, 2013; Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007) but for a number of countries in Europe updates were made drawing on national information provided within EU consultations (Amann et al., 2015) and recent measurements in China where 100,000s of such installations are used in both residential as well as industrial sector (Wang et al., 2009).

Field Code Changed

GAINS distinguishes also open fireplaces as a separate category which is of relevance mostly in North America and some European countries, even though in Europe less than 5% of fuelwood would be used in such installations (Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007).

Here we summarize the published measurements of emission factors for cooking and heating stoves boilers and compare them to the current ranges of region- and technology-specific GAINS values. The focus is on studies that appeared after the original development of the GAINS particulate matter module (Klimont et al., 2002; Kupiainen and Klimont, 2004, 2007).

**Table S2.1:** Summary of PM emission factors for residential wood boilers.

| Emission factors (mg/MJ) | | | | Shares (%) | | | References |
|---|---|---|---|---|---|---|---|
| PM | TC[a] | BC | OC | TC[a] | BC | OC | |
| wood log | | | | | | | |
| 1300<br>(350-2200)[b] | 715 | | | 55 | | | (Boman et al., 2008)<br>old, no accumulator, large fuel charge |
| 120<br>(73-260)[b] | 60 | | | 50 | | | (Boman et al., 2008)<br>old, no accumulator, adjusted fuel charge |
| 95<br>(87-100)[b] | 48 | | | 50 | | | (Boman et al., 2008)<br>old, with accumulator |
| 44<br>(11-450)[b] | 18 | | | 42 | | | (Boman et al., 2008)<br>modern, with accumulator |
| 37 | 27 | 12 | 16 | 75 | 32 | 43 | (Gaegauf et al., 2005), 35 kW apartment house |
| 70-700 | | 20 | 30-335 | | | | GAINS [c], >50 kW, uncontrolled boiler |
| 230-1300 | | 75-200 | 75-600 | | | | GAINS [c], <50 kW, old uncontrolled boiler |
| 80-520 | | 32-50 | 22-230 | | | | GAINS [c], <50 kW, improved |
| 40-260 | | 13-37 | 12-100 | | | | GAINS [c], <50 kW, new/modern |
| wood chip | | | | | | | |
| | | | | 44 | 23 | 21 | (Schmidl et al., 2011) 40 kW moving grate, start-up |
| | | | | 5 | 1 | 4 | (Schmidl et al., 2011) 40 kW moving grate, full load |
| | | | | 35 | 33 | 2 | (Schmidl et al., 2011) 40 kW moving grate, part load |
| 85 | 8 | 2 | 6 | 9 | 2 | 7 | (Gaegauf et al., 2005) 70 kW, institute building |
| wood pellet | | | | | | | |
| 20 | | 0.1 | 0.9 | | 0.5 | 5 | (Lamberg et al., 2011a) efficient combustion |
| 12<br>(3-29)[b] | | 0.8<br>(0-14)[b] | 0.3<br>(0-3)[b] | | 6<br>(0-51)[b] | 2<br>(2-11)[b] | (Lamberg et al., 2011b), 25 kW, nominal load |
| 16 | | 1 | 0.1 | | | | (Tissari et al., 2008), 20 kW, nominal load |
| 24 | | 3 | 0.2 | | | | (Tissari et al., 2008), 20 kW, partial load |
| 49 | 35 | 24 | 11 | 72 | 49 | 23 | (Gaegauf et al., 2005) 10-32 kW, apartment house |
| 8-25 | | 0.8-1 | 0.4-1 | | | | GAINS [c], >50 kW |
| 20-68 | | 5 | 2.5-10 | | | | GAINS [c], <50 kW |

[a] Total Carbon (TC)
[b] (min-max)
[c] PM value refers to PM2.5

**Table S2.2:** Summary of PM emission factors for residential heating wood stoves.

| Emission factors (mg/MJ) | | | Shares (%) | | Reference |
|---|---|---|---|---|---|
| PM | BC | OC | BC | OC | |
| traditional | | | | | |
| 673-1373 | 24-72 | 263-623 | 2-7 | 39-53 | (Alves et al., 2011) |
| 300-1400 | - | - | 2-9 | 35-50 | (Gonçalves et al., 2011) incl. cold start |
| 90-900 | - | - | 2-9 | 35-48 | (Gonçalves et al., 2011) incl. hot start |
| 750-1060 | - | - | - | - | (Jordan and Seen, 2005), full airflow |
| 1560-1700 | - | - | - | - | (Jordan and Seen, 2005), half airflow |
| 1870-3000 | - | - | - | - | (Jordan and Seen, 2005), closed airflow |
| 128-400 | 20 | 157 | 8 | 64 | (McDonald et al., 2000) |
| - | 39-43 | 70-390 | 5-14 | 47-67 | Studies in Kupiainen& Klimont (2007) |
| 150[a] - 930 (2400)[b] | 32[a] - 100 | 60[a] - 435 (1200)[b] | 4-22 | 41-50 | GAINS; the PM value represents PM2.5 |
| improved | | | | | |
| 22-180 | - | - | - | - | (Boman et al., 2008) |
| 86-105 | 9-11 | 52-58 | - | - | (Fine et al., 2004) |
| 130 | 88 | 39 | 68 | 30 | (Gaegauf et al., 2005) |
| 60-160 | - | - | 11-37 | 20-43 | (Gonçalves et al., 2010) |
| 75-97 | 15-28 | 17-35 | 24-32 | 27-39 | (Schmidl et al., 2011) |
| 38-350 | - | - | - | - | (Pettersson et al., 2011) |
| - | 56-79 | 11-16 | - | - | Studies in Kupiainen& Klimont (2007) |
| 55[a] - 372 | 30[a] - 95 | 11[a] -133 | 25-55 | 19-35 | GAINS; the PM value represents PM2.5 |
| new | | | | | |
| 67-122 | 13-15 | 43-67 | - | - | (Fine et al., 2004), catalytic |
| 72-89 | 21-33 | 16-32 | 30-37 | 22-36 | (Schmidl et al., 2011) |
| 30[a] - 186 | 9[a] - 30 | 8[a] - 67 | 18-30 | 28-35 | GAINS; the PM value represents PM2.5 |
| pellet | | | | | |
| 10-66 | - | - | - | - | (Boman et al., 2008) |
| 15-47 | - | - | - | - | (Boman et al., 2011) |
| 17 | 0.7 | - | 4 | - | (Frey et al., 2014) |
| 20 | 0.1 | 0.9 | 0.5 | 5 | (Lamberg et al., 2011b) |
| 3-29 | 0-14 | 0.1-3 | 0-51 | 2-11 | (Lamberg et al., 2011a) |
| - | - | - | 14 | 11 | (Schmidl et al., 2011) |
| 47-129 | 0.5-1.3 | 0.3-5.2 | 1-2 | 1-9 | (Sippula et al., 2007) |
| 10[a] - 47 | 1.3[a] - 4 | 2[a] - 7 | 10-17 | 12-17 | GAINS; the PM value represents PM2.5 |

[a] The lowest values represent Swiss data

[b] Norwegian wood stove

**Table S2.3:** Summary of PM emission factors for cookstoves using biofuels.

| | Emission factors (mg/MJ) | | | References |
|---|---|---|---|---|
| | PM | BC | OC | |
| traditional | | | | |
| | 530 | 44 | 250 | (Just et al., 2013) |
| | 106 | 50 | 44 | (Roden et al., 2009), 3-stone, lab measurements |
| | 515<br>(300-1000) [a] | 83<br>(10-210) [a] | 254<br>(90-660) [a] | (Roden et al., 2009), Honduras, field measurements |
| | 510<br>(280-510) [b] | 65-75<br>(40-75) [b] | 229<br>(125-229) [b] | GAINS [c] |
| improved | | | | |
| | 150 | 80 | 20 | (Just et al., 2013), rocket stove |
| | 270 (100-500) [a] | | | (Li et al., 2009), improved stoves, PM2.5 |
| | 394<br>(120-700) [a] | 102<br>(6-325) [a] | 208<br>(60-460) [a] | (Roden et al., 2009), improved no chimney, field measurements |
| | 205<br>(105-270) [b] | 50-75<br>(27-75) [b] | 63<br>(31-68) [b] | GAINS [c] |
| new | | | | |
| | 255<br>(40-720) [a] | 116<br>(6-660) [a] | 93<br>(33-370) [a] | (Roden et al., 2009), improved with chimney, field measurements |
| | 56-102 | 11-21 | 19-34 | GAINS [c] |
| fan assisted | | | | |
| | 86<br>(25-125) [a] | 33<br>(6-100) [a] | 38<br>(4-71) [a] | (Roden et al., 2009), fan assisted, lab measurements |
| | 54 | 33 | 14 | (Just et al., 2013), gasifier with fan |
| | 17 | 4 | 9 | GAINS [c] |

[a] (min-max)

[b] central value for fuelwood and in brackets the whole range including also dung and agricultural residues

[c] the PM value represents PM2.5

**Table S2.4:** Summary of PM emission factors for coal cooking and heating stoves

| Emission factors (mg/MJ) | | | References |
|---|---|---|---|
| PM | BC | OC | |
| traditional | | | |
| 805 (214-1360) [a] | 250 (11-540) [a] | 400 (116-710) [a] | (Zhi et al., 2009), portable stove, bituminous coals |
| | 332 (10-610) [a] | 472 (129-822) [a] | (Chen et al., 2009), simple low-efficiency stove without chimney, bituminous coals |
| 351 | 135 | 108 | GAINS [b] (cooking) |
| 315-495 | 90-220 | 160-200 | GAINS [b] (heating) |
| improved | | | |
| | 466 (6-1377) [a] | 248 (35-551) [a] | (Chen et al., 2009), high-efficiency stove with chimney |
| 492 | 183 | 200 | (Zhang et al., 2008), steel stove, brown coal |
| 36 | 1 | 16 | (Zhang et al., 2008), steel stove, bituminous coal |
| 408 (155-685) [a] | 40 (2-140) [a] | 230 (78-470) [a] | (Zhi et al., 2009), bituminous coals |
| 246 | 132 | 60 | GAINS [b] (cooking) |
| 315-350 | 82-200 | 88-112 | GAINS [b] (heating) |
| new | | | |
| 270 | 23 | 96 | (Li et al., 2016), average for bituminous coals |
| 176 | 108 | 32 | GAINS [b] (cooking) |
| 158-248 | 73-176 | 48-60 | GAINS [b] (heating) |
| briquettes | | | |
| | 16 (2-33) [a] | 329 (71-668) [a] | (Chen et al., 2009), simple low-efficiency, no chimney |
| | 4 (0.5-9) [a] | 219 (27-423) [a] | (Chen et al., 2009), high-efficiency, with chimney |
| 184 | 3 | 80 | (Zhang et al., 2008), steel stove |
| 440 (98-930) [a] | 12 (2-23) [a] | 233 (67-460) [a] | (Zhi et al., 2009), traditional portable stove |
| 202 (90-346) [a] | 2 (0.5-6) [a] | 124 (36-217) [a] | (Zhi et al., 2009), improved stove with chimney |
| 17 | 0.4 | 6.5 | (Li et al., 2016), semi-coke briquettes |
| 23-135 | 0.3-1 | 9-55 | GAINS [b] |

[a] (min-max)

[b] the PM value represents PM2.5

## S3 Summary of particulate matter emissions factors for diesel generators

Note that the ranges presented for GAINS represent the spread across GAINS regions or technologies (if a category refers to an aggregate across several measures) defined in the GAINS model.

**Table S3.1:** Summary of PM emission factors for diesel generator sets

| Emission factors (mg/MJ) | | | Shares (%) | | Reference |
|---|---|---|---|---|---|
| PM | BC | OC | BC | OC | |
| 69-189 | | | | | Uma et al. (2004), 10 kW (higher value), 40 kW (lower value) |
| 139 | | | 66% | | Bond et al. (2004) |
| 13/22 | | | | | Gilmore et al. (2006), ICE 10 kW, with/without DPF |
| | | 116-585 | | | Watson et al. (2006) [a] |
| 59-190 | 12-54 | 30-120 | 31% | 51% | Shah et al. (2007) [b] 300 kW 1985 Detroit Diesel V92, 2-str |
| 45-219 | 30-145 | 8-56 | 67% | 21% | Shah et al. (2007) [b] 350 kW 2000 Cat 3406C, 4-str |
| 22-143 | 10-80 | 6-37 | 53% | 25% | Shah et al. (2007) [b] 300 kW 1985 Detroit Diesel V92, 2-str, DOC |
| 59-203 | 28-145 | 4-16 | 67% | 8% | Shah et al. (2007) [b] 350 kW 2000 Cat 3406C, 4-str, DOC |
| 23-190 | 9-96 | 10-81 | 49% | 36% | Shah et al. (2007) [b] 300 kW 1985 Detroit Diesel V92, 2-str, DOC+FBC |
| 4-26 | 2.5-19 | 1-3 | 76% | 15% | Shah et al. (2007) [b] 350 kW 2000 Cat 3406C, 4-str, passive-DPF |
| 1-3 | 0.8-2 | 1-6 | 67% | 49% | Shah et al. (2007) [b] 350 kW 2000 Cat 3406C, 4-str, active-DPF |
| | | | | 20-70% | Watson et al. (2008) |
| 14-42 | | | | | Zhu et al. (2009) [c] |
| 174-433 | | | | | Tsai et al. (2010) [d] |
| 55 | | | | | Anayochukwu et al. (2013) |
| GAINS emission factors; the PM value represents PM2.5 | | | | | |
| 96 | 40 | 28 | 41% | 29% | No control |
| 48-64 | 20-26 | 14-19 | 41% | 29% | Controlled, no DPF |
| <1-3 | 0.5-2 | 0.3-0.8 | | | Controlled, with DPF |

[a] Higher value with 10% load and lower value with 100% load for a 100 kW DG set

[b] Lower value with 100% load and higher value with 10% load, share of BC/OC is average of all loads

[c] Average of 14 military diesel generators with rated capacities of 10, 30, 60, and 100 kW under different load conditions. The fleet average EFs are 1.2+/-0.6 g/kg for PM.

[d] Higher value with no load and lower value with 10 kW

## S4 Transport sector

Note that the ranges presented for GAINS represent the spread across GAINS regions or technologies (if a category refers to an aggregate across several measures) defined in the GAINS model.

**Table S4.1:** Comparison of selected measured emissions factors and ranges used in the GAINS model for diesel and gasoline cars and light duty vehicles.

| | Emission factors (mg/MJ) | | | Reference |
|---|---|---|---|---|
| | PM | BC | OC | |
| Diesel | | | | |
| Pre-/early regulation | 44-67 | 9-17 | 13-34 | (Subramanian et al., 2009) |
| Euro 1 | 67 | 17 | 13 | (Subramanian et al., 2009) |
| Euro 2 | 30-33 | 7-16 | 8-12 | (Cheung et al., 2009; Subramanian et al., 2009) |
| Euro 3 | 10-29 | | | (Graham, 2005) |
| Euro 4 | 6-11 | 3-8 | 1-2 | (Cheung et al., 2009; Geller et al., 2006) |
| Euro 4 with DPF | 0.2-0.3 | 0.02-0.1 | 0.02-0.06 | (Dwyer et al., 2010; Louis et al., 2016) |
| Pre-/early regulation | 56-133 | 38-76 | 21-51 | GAINS [a] |
| Euro 1 | 22-50 | 16-35 | 5-11 | GAINS [a]; for developing countries the values only marginally lower than pre/early regulation |
| Euro 2 | 15-40 | 12-32 | 3-6 | GAINS [a]; for developing countries the values only marginally lower than pre/early regulation |
| Euro 3 | 11-29 | 10-22 | 1-2 | GAINS [a] |
| Euro 4 | 5-20 | 4-17 | 0.5-1.6 | GAINS [a] |
| Euro 4 with DPF | 0.5-1 | 0.1-0.3 | 0.1-0.6 | GAINS [a] |
| Gasoline | | | | |
| Pre-/early regulation | 4-10 | 0.5-2 | 2-10 | see studies in Kupiainen and Klimont (2004, 2007) |
| Euro 1, 2 | 1-4 | 0.6-1.5 | 0.3-1.6 | see studies in Kupiainen and Klimont (2004, 2007) |
| Euro 3 | 0.2-2 | 0.01-0.2 | 0.2-0.6 | (Cheung et al., 2009; Geller et al., 2006; Graham, 2005) |
| Euro 4 | | 0.001-0.4 | | (Louis et al., 2016) |
| Pre-/early regulation | 6 | 1 | 3-4 | GAINS [a] |
| Euro 1, 2 | 1-4 | 0.2-1 | 0.3-1.7 | GAINS [a] |
| Euro 3, 4 | 0.3-1.1 | 0.05-0.5 | 0.1-0.4 | GAINS [a] |

[a] the PM value represents PM2.5

**Table S4.2:** Comparison of selected measured emissions factors and ranges used in the GAINS model for diesel heavy duty vehicles

| | Emission factors (mg/MJ) | | | Reference |
|---|---|---|---|---|
| | PM | BC | OC | |
| Diesel heavy duty trucks | | | | |
| Pre-/early regulation | 28-33 | | | (Herner et al., 2009; Yanowitz et al., 2000) |
| | 44-244 | 4-50 | 15-122 | (Subramanian et al., 2009), Bangkok, Thailand |
| | 30-50 | | | (Liu et al., 2009), on-road measurements in China |
| Euro I | 11 | | | (Yanowitz et al., 2000) |
| | 22 | 4 | 9 | (Subramanian et al., 2009), Bangkok, Thailand |
| | 10-20 | | | (Liu et al., 2009), on-road measurements in China |
| Euro II | 22-44 | 2-9 | 7-22 | (Subramanian et al., 2009), Bangkok, Thailand |
| | 7-17 | 16 | | (Liu et al., 2009; Wang et al., 2011), on-road measurements in China |
| Euro III | 3-7 | 9 | | (Liu et al., 2009; Wang et al., 2011), on-road measurements in China |
| Euro IV | | 4 | | (Wang et al., 2011), on-road measurements in China |
| Pre-/early regulation | 34-107 | 17-53 | 10-37 | GAINS [a] |
| Euro I | 21-71 | 17-53 | 6-19 | GAINS [a] |
| Euro II | 11-44 | 7-30 | 2-10 | GAINS [a] |
| Euro III | 10-27 | 8-25 | 2-7 | GAINS [a] |
| Euro IV, V | 2-7 | 2-5 | 0.3-1 | GAINS [a] |
| Euro VI | 0.1-0.4 | 0.01-0.06 | 0.06-0.15 | GAINS [a] |

[a] the PM value represents PM2.5

**Table S4.3:** Comparison of selected measured emissions factors and ranges used in the GAINS model for non-road machinery.

| | Emission factors (mg/MJ) | | | Reference |
|---|---|---|---|---|
| | PM | BC | OC | |
| Diesel locomotives | | | | |
| Pre-/early regulation | 49-67 | | | (Dincer and Elbir, 2007; Johnson et al., 2013; Tang et al., 2015) |
| Regulated | 20-40 | 20 | | (Dincer and Elbir, 2007; Galvis et al., 2013; Johnson et al., 2013; Tang et al., 2015) |
| | 30 | 14 | | (Galvis et al., 2013) |
| | 20 | 15 | | (Jaffe et al., 2014) |
| | 37 | 21 | | (Krasowsky et al., 2015) |
| pre-regulated | 49-98 | 24-45 | 12-25 | GAINS [a] |
| regulated (stage I) | 26-49 | 11-22 | 6-12 | GAINS [a] |
| Agriculture | | | | |
| Pre-regulation | 141 | 58 | 41 | (Kupiainen and Klimont, 2007) |
| | 89 | 49 | | (EEA, 2013) |
| Stage I | 20-39 | 16-21 | | (EEA, 2013) |
| Stage II | 15 | 11.5 | | (EEA, 2013) |
| Pre-regulation | 100-170 | 41-70 | 29-50 | GAINS [a] |
| Stage I | 57-96 | 23-40 | 16-27 | GAINS [a] |
| Stage II, III | 27-43 | 10-19 | 8-12 | GAINS [a] |
| Stage IV,V | 6-10 | 0.7-1.2 | 0.5-0.8 | GAINS [a] |
| Construction | | | | |
| Pre-regulation | 140 | 65 | 30 | (Kupiainen and Klimont, 2007) |
| | 103 | 56 | | (EEA, 2013) |
| Stage I | 85 | 47 | | (EEA, 2013) |
| Pre-regulation | 95-140 | 46-68 | 21-31 | GAINS [a] |
| Stage I | 57-76 | 26-39 | 12-18 | GAINS [a] |
| Stage II, III | 24-36 | 12-17 | 5-8 | GAINS [a] |
| Stage IV,V | 6-8 | 0.8-1.2 | 0.4-0.6 | GAINS [a] |

[a] the PM value represents PM2.5

**Table S4.4:** Comparison of selected measured emissions factors and ranges used in the GAINS model for 2-wheelers.

| | Emission factors (mg/MJ) | | | References |
|---|---|---|---|---|
| | PM | BC | OC | |
| 2-stroke | | | | |
| Euro 0 mopeds | 250 (198-295) | | | (Spezzano et al., 2008), hot start |
| | 160 (121-878) | | | (Spezzano et al., 2008), cold start |
| Euro 1 mopeds | 169 (102-235) | | | (Spezzano et al., 2008), hot start |
| | 42 (26-71) | | | (Spezzano et al., 2008), cold start |
| Euro 2 mopeds | 147-217 | | | (Spezzano et al., 2008), hot start |
| | 13-215 | | | (Spezzano et al., 2008), cold start |
| CNG rickshaw, Delhi, India | 124-160 | | | (Grieshop et al., 2012) |
| Euro 0 mopeds | 132-1400 | 10-75 | 90-1015 | GAINS [a] |
| Euro 1 mopeds | 12-450 | 7-49 | 40-300 | GAINS [a] |
| Euro 2 mopeds | 37-280 | 6-45 | 23-172 | GAINS [a] |
| Euro 3 mopeds | 14-112 | 3-30 | 8-61 | GAINS [a] |
| 4-stroke | | | | |
| Motorcycles | 2.6-3.7 | | | (Yang et al., 2005), cold start |
| Euro 0 motorcycles | 4 | | | (Spezzano et al., 2007) |
| Euro 1 motorcycles | 2 | | | (Spezzano et al., 2007) |
| Rickshaw, Delhi, India | 30-45 | | | (Grieshop et al., 2012) |
| CNG rickshaw, Delhi, India | 12-13 | | | (Grieshop et al., 2012) |
| Euro 0 motorcycles | 6-14 | 1-2 | 3-9 | GAINS [a] |
| Euro 1 motorcycles | 5-12 | 1-2 | 2-7 | GAINS [a] |
| Euro 2 motorcycles | 3-5 | 0.5-0.8 | 0.4-1.7 | GAINS [a] |
| Euro 3 motorcycles | 2-3 | 0.5-0.75 | 0.3-1.4 | GAINS [a] |

[a] the PM value represents PM2.5

**Table S4.5:** Summary of PM emission factor ranges used in the GAINS model for non-exhaust transport sources

| | Emission factors (mg/km) | | | |
|---|---|---|---|---|
| | $PM_{10}$ | $PM_{2.5}$ | BC | OC |
| Brake wear | | | | |
| Cars | 3.5 – 12 | 2.5 – 5 | 0.05 – 0.12 | 0.8 – 2.2 |
| Light duty vehicles | 3.5 – 19 | 2.5 – 8 | 0.05 – 0.2 | 0.8 – 3.5 |
| Heavy duty vehicles | 21 – 53 | 13 – 21 | 0.25 – 0.5 | 5 – 17 |
| Tyre wear | | | | |
| Cars | 1.5 – 9 | 0.15 – 0.7 | 0.2 – 1 | 0.5 - 2.4 |
| Light duty vehicles | 2.5 – 7 | 0.2 – 0.7 | 0.35 – 1 | 0.85 – 2.4 |
| Heavy duty vehicles | 40 – 47 | 4.2 – 4.7 | 6 – 7 | 15 – 17 |
| Road abrasion | | | | |
| Cars & Light duty vehicles | 7 - 10 | 3 – 5 | 0.15 – 0.6 | 0.7 – 1 |
| | 30 – 140 [a] | 20 – 80 [a] | 0.2 – 1.5 [a] | 4 – 14 [a] |
| Heavy duty vehicles | 38 – 50 | 18 – 27 | 0.7 – 1 | 3 – 5 |

[a] vehicles with studded tires; variation between estimates for Scandinavian and alpine countries

## S5 Industry

GAINS model PM emission factors (as used for the ECLIPSE *V5a*) for brick making compared with values used in GAINS previously (UNEP/WMO, 2011) and recent set of measurements on typical kilns used in South Asia (Weyant et al., 2014).

**Table S5.1:** Comparison of emissions factors used in the GAINS model for brick kilns with selected other studies.

| Emission factors (g kg$^{-1}$ brick) | | | References |
|---|---|---|---|
| PM2.5 | BC | OC | |
| Clamp kiln | | | |
| 1.6 | 0.35 | 0.3 | (UNEP/WMO, 2011)[a] |
| 1 | 0.3 | 0.1 | GAINS (Asia) |
| 1 | 0.35 | 0.15 | GAINS (Latin America and Africa) |
| Downdraft kiln | | | |
| 0.49 | 0.19 | 0.07 | (Weyant et al., 2014) |
| 0.97 | 0.29 | 0.09 | GAINS (all regions) |
| Bull’s trench kiln (BTK) | | | |
| 1.31 | 0.27 | 0.24 | (UNEP/WMO, 2011)[a] |
| 0.19 (0.08-0.33) | 0.15 (0.09-0.27) | 0.007 | (Weyant et al., 2014)[b] |
| 0.18/0.8 | 0.13/0.25 | 0.01/0.07 | GAINS (Asia); fixed /moving chimney |
| Vertical shaft brick kiln (VSBK) | | | |
| 0.77 | 0.175 | 0.15 | (UNEP/WMO, 2011)[a] |
| 0.07 (0.005-0.009) | 0.0015 (0.001-0.002) | 0.014 | (Weyant et al., 2014)[b] |
| 0.093 | 0.001-0.004 | 0.002-0.059 | GAINS (Asia) |
| 0.093 | 0.002 | 0.059 | GAINS (Latin America and Africa) |
| Zig-zag kiln | | | |
| 0.06 (0.03-0.06) | 0.01 (0.014-0.03) | 0.005 | (Weyant et al., 2014)[b] |
| 0.13 | 0.04 | 0.02 | GAINS (Asia) |
| Tunnel kiln (coal) | | | |
| 0.28 | 0.0035 | 0.003 | (UNEP/WMO, 2011)[a] |
| 0.24 | 0.001 | <0.00 | (Weyant et al., 2014) |
| 0.18 | 0.002 | 0.0035 | GAINS (all regions) |
| Hoffman kiln | | | |
| 0.08 | 0.003 | 0.005 | GAINS (all regions) |
| Marquez kiln (MK) | | | |
| 0.15 | 0.06 | 0.02 | GAINS (Latin America) |

[a] Previous version of the GAINS model was used
[b] Central value and ranges of average values; all measurement data provided in the original study

Brick sector production structure in Asia has been analysed in a number of studies addressing either the whole region where selected countries, typically key producers including China, India, Pakistan, Bangladesh, Vietnam, are discussed (AIT, 2003; BASIN, 1999; FAO, 1993; Heierli and Maithel, 2008; Maithel, 2014) or focusing on particular countries like China (Zhang, 1997), India (BASIN, 1998; Maithel et al., 2012; Verma and Uppal, 2013), Bangladesh (Croitoru and Sarraf, 2012; Guttikunda et al., 2013; World Bank, 2011), Cambodia (Rozemuller, 1999), Afghanistan (Samuel Hall Consulting, 2011), Nepal (Heierli et al., 2007).  More recently, a number of development programs and local air pollution studies focused on this sector in the Latin America and Caribbean regions, including some where information about kiln structure was collected (Bellprat, 2009; EELA, 2011; Erbe, 2011; PRAL, 2012; Stratus Consulting, 2014; SwissContact, 2014a). Fewer assessments exist for Africa (Scott, 2013; SwissContact, 2014c). The updated and country specific data for Latin America and Caribbean (LAC) is included only in version *V5a* of ECLIPSE since the previous versions included just five regions for the whole LAC; Argentina, Brazil, Chile, Mexico, other LAC.

GAINS activity data has been built on the basis of several regional studies where production, energy efficiency, and sector structure were discussed, i.e., Asia (AIT, 2003; Co et al., 2009; Croitoru and Sarraf, 2012; FAO, 1993; Guttikunda et al., 2013; Heierli et al., 2007; Heierli and Maithel, 2008; Maithel, 2014; Maithel et al., 2012; Samuel Hall Consulting, 2011; Subrahmanya, 2006; Verma and Uppal, 2013; World Bank, 2011; Zhang, 1997), Africa (Alam, 2006; Scott, 2013; SwissContact, 2014c), Latin America and Caribbean  (Bellprat, 2009; EELA, 2011; PRAL, 2012; Stratus Consulting, 2014; SwissContact, 2014b). For several countries where we found no regional analysis, the United Nations data on 'building bricks, made of clay' was used (http://unstats.un.org/unsd/industry/commoditylist2.asp). There are some differences between different versions of the ECLIPSE datasets; specifically during the development of the *V5a* version, the data for all countries in Latin America and Caribbean was revisited and updated, and a new version of the UN statistics was downloaded.

**Table S5.2:** Brick production in key regions; GAINS model assumptions - ECLIPSE *V5a*, Tg bricks year$^{-1}$

| | 1990 | 1995 | 2000 | 2005 | 2010 |
|---|---|---|---|---|---|
| Global | 1542 | 2357 | 2688 | 3022 | 3574 |
| Asia | 1314 | 2130 | 2530 | 2819 | 3320 |
| *of which:* | | | | | |
| *China* | 1050 | 1800 | 2106 | 2204 | 2508 |
| *India* | 131 | 178 | 254 | 406 | 553 |
| *Vietnam* | 20 | 20 | 27 | 46 | 65 |
| *Bangladesh* | 9 | 15 | 18 | 17 | 25 |
| *Pakistan* | 32 | 41 | 50 | 59 | 74 |
| *Other Asia* | 71 | 75 | 76 | 87 | 95 |
| Africa | 18 | 18 | 15 | 17 | 22 |
| Europe | 158 | 156 | 72 | 82 | 79 |
| Latin America and Caribbean | 29 | 30 | 43 | 75 | 127 |
| Other | 23 | 23 | 27 | 29 | 25 |

### S6 Emissions of PM species over time in ECLIPSE datasets

The Fig S6.1 shows emissions of PM10, PM2.5, BC, and OC calculated with the GAINS model within different versions of the ECLIPSE dataset. These have been created between 2013 and 2015 and include a number of updates to activity data and emission factors; the methodology remained the same. The changes for PM10 and PM2.5 are similar, driven by updates of activity data, i.e., the energy statistics from IEA were reimported for the whole time series for the version *V5* and *V5a* and for China the regional coal statistics were used. Control strategies have been updated continuously considering more up to date information available over time. Additionally, in version *V5a* Latin America and Caribbean were revised since higher spatial resolution was introduced in the GAINS model. Several of the above mentioned updates affected also emissions of BC and OC but the largest impact on the BC emissions was due to introduction of emissions from kerosene lamps which were not specifically distinguished in *V4a*; this represents the key component of the higher emissions in *V5*, *V5a*. For OC the change is in the opposite direction and *V5a* has significantly lower emissions than previous versions which is due to update of the OC emission factor for residential cooking in Asia and Africa.

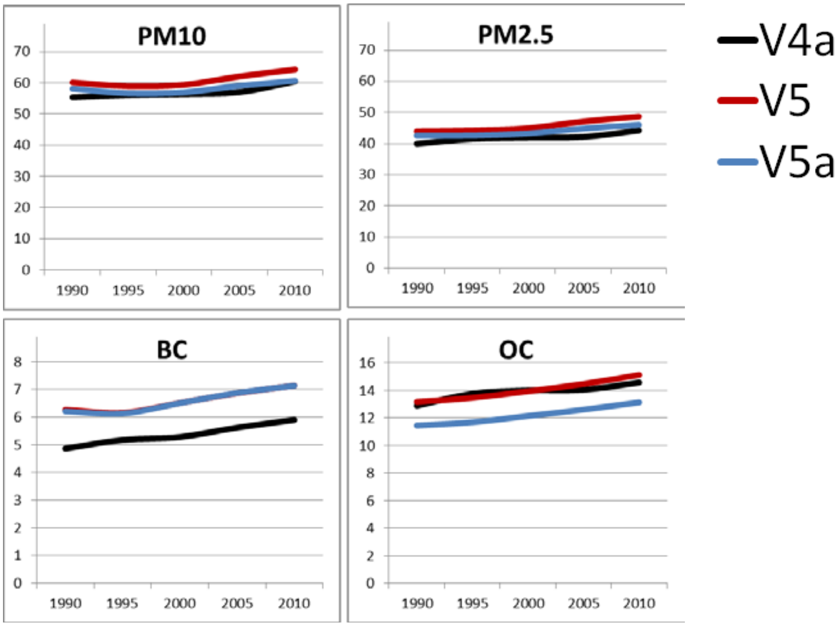


**Figure S6.1.** Global emissions of PM (excluding international shipping and open biomass burning) in the period 1990-2010 in different ECLIPSE scenarios; unit [Tg year$^{-1}$]

**Table S6.2:** Global anthropogenic (excluding international shipping & aviation) emissions of PM10 in ECLIPSE V5a; [Gg year$^{-1}$]

| Region | 1990 | 1995 | 2000 | 2005 | 2010 |
|---|---|---|---|---|---|
| 1 Canada | 333 | 315 | 345 | 337 | 334 |
| 2 USA | 2416 | 2158 | 1954 | 1920 | 1630 |
| 3 Mexico | 643 | 621 | 653 | 574 | 572 |
| 4 Rest Central America | 454 | 455 | 479 | 498 | 516 |
| 5 Brazil | 1228 | 1295 | 1250 | 1385 | 1456 |
| 6 Rest South America | 1018 | 1155 | 1138 | 1131 | 1192 |
| 7 Northern Africa | 1022 | 1152 | 1355 | 1144 | 1194 |
| 8 Other Africa | 4393 | 4993 | 5831 | 6425 | 7150 |
| 10 South Africa | 682 | 738 | 747 | 848 | 818 |
| 11 Western Europe | 3294 | 2458 | 2031 | 1747 | 1577 |
| 12 Central Europe | 2944 | 1608 | 1236 | 1046 | 1038 |
| 13 Turkey | 1007 | 756 | 525 | 477 | 571 |
| 14 Ukraine+ | 1854 | 856 | 679 | 707 | 680 |
| 15 Asia-Stan | 836 | 325 | 303 | 314 | 392 |
| 16 Russia+ | 5833 | 2434 | 2314 | 2316 | 2161 |
| 17 Middle East | 836 | 954 | 1055 | 962 | 996 |
| 18 India | 7828 | 8785 | 8654 | 7952 | 8061 |
| 19 Korea | 1227 | 913 | 844 | 816 | 768 |
| 20 China+ | 14057 | 17612 | 18205 | 21230 | 21976 |
| 21 Southeastern Asia | 2291 | 2855 | 2783 | 2451 | 2526 |
| 22 Indonesia+ | 1383 | 1576 | 1673 | 1768 | 1902 |
| 23 Japan | 545 | 435 | 354 | 319 | 267 |
| 24 Oceania | 295 | 303 | 354 | 354 | 342 |
| 25 Rest South Asia | 1695 | 1894 | 2211 | 2349 | 2533 |
| **Global** | **58112** | **56646** | **56974** | **59071** | **60651** |

**Table S6.3:** Global anthropogenic (excluding international shipping & aviation) emissions of PM2.5 in ECLIPSE V5a; [Gg year$^{-1}$]

| Region | 1990 | 1995 | 2000 | 2005 | 2010 |
|---|---|---|---|---|---|
| 1 Canada | 252 | 244 | 250 | 242 | 241 |
| 2 USA | 1629 | 1482 | 1296 | 1275 | 1027 |
| 3 Mexico | 495 | 498 | 526 | 459 | 454 |
| 4 Rest Central America | 395 | 394 | 416 | 428 | 446 |
| 5 Brazil | 938 | 974 | 933 | 1054 | 1098 |
| 6 Rest South America | 825 | 933 | 909 | 901 | 949 |
| 7 Northern Africa | 762 | 852 | 982 | 847 | 909 |
| 8 Other Africa | 4056 | 4606 | 5308 | 5887 | 6575 |
| 10 South Africa | 408 | 444 | 431 | 501 | 490 |
| 11 Western Europe | 2125 | 1700 | 1360 | 1157 | 1037 |
| 12 Central Europe | 1610 | 1020 | 843 | 752 | 775 |
| 13 Turkey | 585 | 480 | 388 | 356 | 425 |
| 14 Ukraine+ | 1072 | 531 | 464 | 483 | 455 |
| 15 Asia-Stan | 562 | 222 | 211 | 222 | 283 |
| 16 Russia+ | 3702 | 1614 | 1530 | 1495 | 1413 |
| 17 Middle East | 686 | 778 | 845 | 784 | 794 |
| 18 India | 5768 | 6453 | 6472 | 5957 | 6032 |
| 19 Korea | 784 | 600 | 547 | 565 | 529 |
| 20 China+ | 10863 | 13072 | 13633 | 15673 | 16096 |
| 21 Southeastern Asia | 1878 | 2257 | 2198 | 1974 | 2012 |
| 22 Indonesia+ | 1230 | 1371 | 1447 | 1510 | 1604 |
| 23 Japan | 337 | 295 | 236 | 203 | 160 |
| 24 Oceania | 188 | 193 | 210 | 201 | 188 |
| 25 Rest South Asia | 1455 | 1629 | 1859 | 1962 | 2065 |
| **Global** | **42606** | **42640** | **43294** | **44888** | **46055** |

**Table S6.4:** Global anthropogenic (excluding international shipping & aviation) emissions of PM1 in ECLIPSE V5a; [Gg year$^{-1}$]

| Region | 1990 | 1995 | 2000 | 2005 | 2010 |
|---|---|---|---|---|---|
| 1 Canada | 184 | 195 | 196 | 187 | 190 |
| 2 USA | 1163 | 1095 | 949 | 930 | 727 |
| 3 Mexico | 375 | 378 | 395 | 361 | 357 |
| 4 Rest Central America | 329 | 331 | 353 | 366 | 390 |
| 5 Brazil | 706 | 720 | 718 | 819 | 846 |
| 6 Rest South America | 657 | 732 | 708 | 712 | 764 |
| 7 Northern Africa | 447 | 476 | 514 | 485 | 542 |
| 8 Other Africa | 3724 | 4213 | 4838 | 5416 | 6064 |
| 10 South Africa | 285 | 309 | 307 | 354 | 354 |
| 11 Western Europe | 1397 | 1171 | 966 | 834 | 751 |
| 12 Central Europe | 894 | 667 | 619 | 579 | 607 |
| 13 Turkey | 386 | 341 | 286 | 263 | 311 |
| 14 Ukraine+ | 565 | 325 | 279 | 278 | 261 |
| 15 Asia-Stan | 292 | 154 | 146 | 154 | 198 |
| 16 Russia+ | 1988 | 1078 | 1011 | 936 | 852 |
| 17 Middle East | 501 | 562 | 596 | 614 | 615 |
| 18 India | 4500 | 4992 | 5016 | 4700 | 5031 |
| 19 Korea | 635 | 510 | 450 | 464 | 429 |
| 20 China+ | 9153 | 11251 | 11731 | 12473 | 11606 |
| 21 Southeastern Asia | 1800 | 2204 | 2093 | 1791 | 1803 |
| 22 Indonesia+ | 1135 | 1254 | 1315 | 1373 | 1453 |
| 23 Japan | 258 | 229 | 157 | 126 | 87 |
| 24 Oceania | 140 | 146 | 155 | 143 | 133 |
| 25 Rest South Asia | 1303 | 1445 | 1625 | 1714 | 1811 |
| **Global** | **32816** | **34780** | **35422** | **36073** | **36180** |

**Table S6.5:** Global anthropogenic (excluding international shipping & aviation) emissions of BC in ECLIPSE V5a; [Gg year$^{-1}$]

| Region | 1990 | 1995 | 2000 | 2005 | 2010 |
|---|---|---|---|---|---|
| 1 Canada | 44 | 49 | 51 | 49 | 49 |
| 2 USA | 311 | 291 | 281 | 279 | 201 |
| 3 Mexico | 76 | 77 | 82 | 84 | 88 |
| 4 Rest Central America | 52 | 54 | 61 | 65 | 71 |
| 5 Brazil | 143 | 148 | 160 | 171 | 179 |
| 6 Rest South America | 115 | 135 | 140 | 150 | 169 |
| 7 Northern Africa | 127 | 120 | 117 | 121 | 140 |
| 8 Other Africa | 752 | 836 | 942 | 1030 | 1135 |
| 10 South Africa | 57 | 59 | 57 | 74 | 72 |
| 11 Western Europe | 331 | 335 | 307 | 287 | 246 |
| 12 Central Europe | 126 | 112 | 112 | 121 | 134 |
| 13 Turkey | 60 | 59 | 53 | 51 | 67 |
| 14 Ukraine+ | 88 | 59 | 45 | 41 | 36 |
| 15 Asia-Stan | 50 | 28 | 33 | 38 | 55 |
| 16 Russia+ | 439 | 251 | 238 | 226 | 177 |
| 17 Middle East | 174 | 183 | 210 | 243 | 262 |
| 18 India | 853 | 931 | 884 | 908 | 1022 |
| 19 Korea | 135 | 84 | 71 | 84 | 74 |
| 20 China+ | 1348 | 1347 | 1655 | 1823 | 1924 |
| 21 Southeastern Asia | 300 | 299 | 304 | 328 | 333 |
| 22 Indonesia+ | 243 | 260 | 275 | 279 | 290 |
| 23 Japan | 67 | 74 | 66 | 50 | 29 |
| 24 Oceania | 30 | 32 | 35 | 35 | 33 |
| 25 Rest South Asia | 288 | 304 | 325 | 337 | 348 |
| **Global** | **6210** | **6129** | **6505** | **6872** | **7134** |

**Table S6.6:** Global anthropogenic (excluding international shipping & aviation) emissions of OC in ECLIPSE V5a; [Gg year$^{-1}$]

| Region | 1990 | 1995 | 2000 | 2005 | 2010 |
|---|---|---|---|---|---|
| 1 Canada | 72 | 77 | 77 | 72 | 74 |
| 2 USA | 448 | 434 | 388 | 379 | 308 |
| 3 Mexico | 162 | 162 | 164 | 158 | 155 |
| 4 Rest Central America | 144 | 149 | 159 | 169 | 181 |
| 5 Brazil | 251 | 258 | 275 | 311 | 314 |
| 6 Rest South America | 297 | 329 | 315 | 324 | 344 |
| 7 Northern Africa | 145 | 150 | 155 | 166 | 192 |
| 8 Other Africa | 1627 | 1842 | 2124 | 2408 | 2701 |
| 10 South Africa | 101 | 108 | 110 | 129 | 130 |
| 11 Western Europe | 495 | 422 | 343 | 284 | 253 |
| 12 Central Europe | 224 | 201 | 217 | 220 | 234 |
| 13 Turkey | 114 | 108 | 95 | 88 | 107 |
| 14 Ukraine+ | 149 | 102 | 82 | 77 | 72 |
| 15 Asia-Stan | 90 | 66 | 62 | 64 | 86 |
| 16 Russia+ | 509 | 332 | 304 | 256 | 231 |
| 17 Middle East | 190 | 217 | 220 | 237 | 229 |
| 18 India | 1530 | 1623 | 1596 | 1630 | 1755 |
| 19 Korea | 200 | 157 | 147 | 157 | 148 |
| 20 China+ | 3147 | 3264 | 3500 | 3564 | 3599 |
| 21 Southeastern Asia | 526 | 548 | 567 | 598 | 632 |
| 22 Indonesia+ | 431 | 473 | 514 | 551 | 595 |
| 23 Japan | 51 | 54 | 49 | 40 | 29 |
| 24 Oceania | 52 | 55 | 57 | 51 | 46 |
| 25 Rest South Asia | 502 | 562 | 628 | 680 | 726 |
| **Global** | **11456** | **11695** | **12150** | **12610** | **13140** |

The following charts in this section show emissions of PM species by key sectors for seven global regions defined as in Figure 3 in the main paper. These charts are the same as Figure 4 in the paper where global emissions are shown. Note that the scale varies across sectors and also across regions. For all regions, except Pacific, the units are million tons [Tg year$^{-1}$]; for Pacific they are [Gg year$^{-1}$].

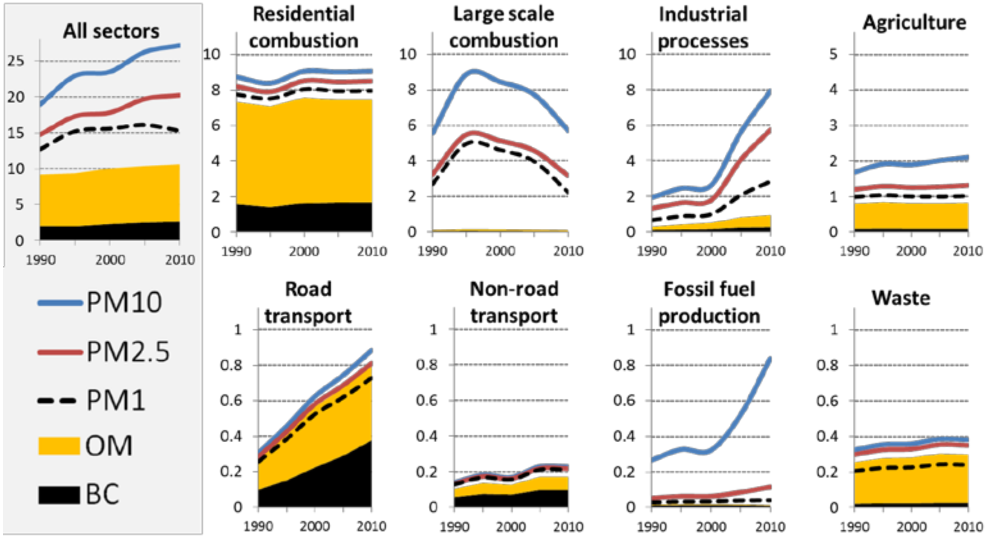


**Figure S6.2.** Emissions of PM species by key sectors in *East Asia* in the period 1990-2010 [Tg year$^{-1}$]; ECLIPSE *V5a*

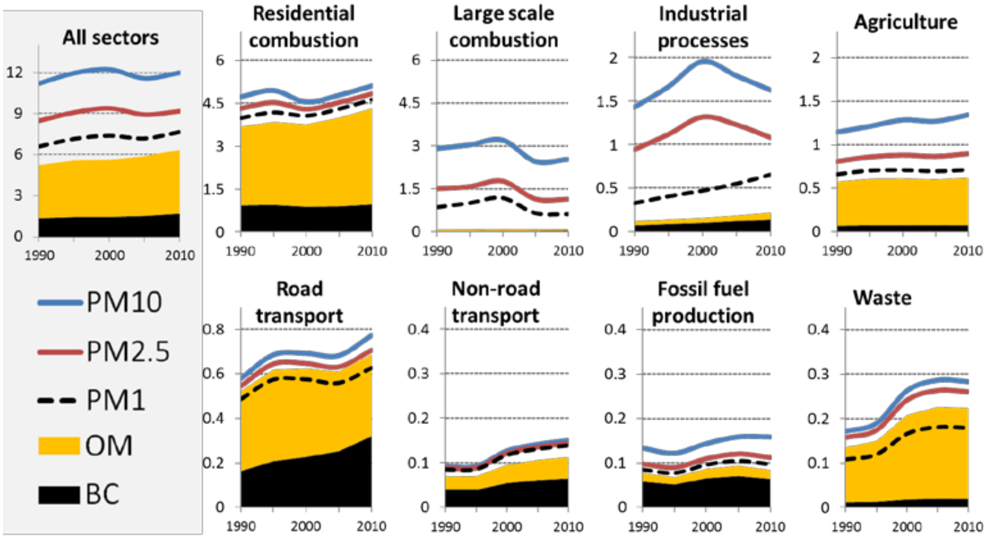


**Figure S6.3.** Emissions of PM species by key sectors in *Other Asia* in the period 1990-2010 [Tg year$^{-1}$]; ECLIPSE *V5a*

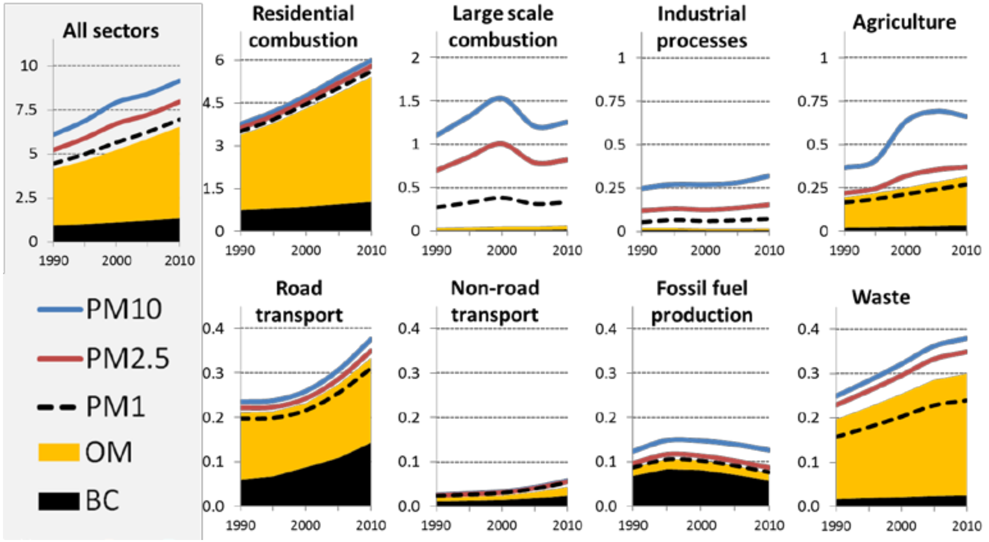


**Figure S6.4.** Emissions of PM species by key sectors in *Africa* in the period 1990-2010 [Tg year$^{-1}$]; ECLIPSE *V5a*

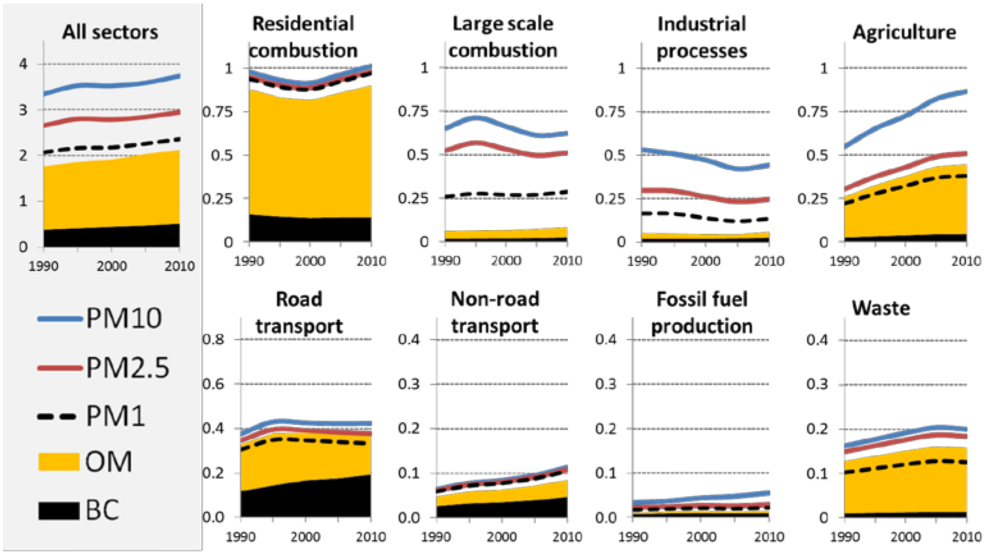


**Figure S6.5.** Emissions of PM species by key sectors in *Latin America and Caribbean* in the period 1990-2010 [Tg year$^{-1}$]; ECLIPSE *V5a*

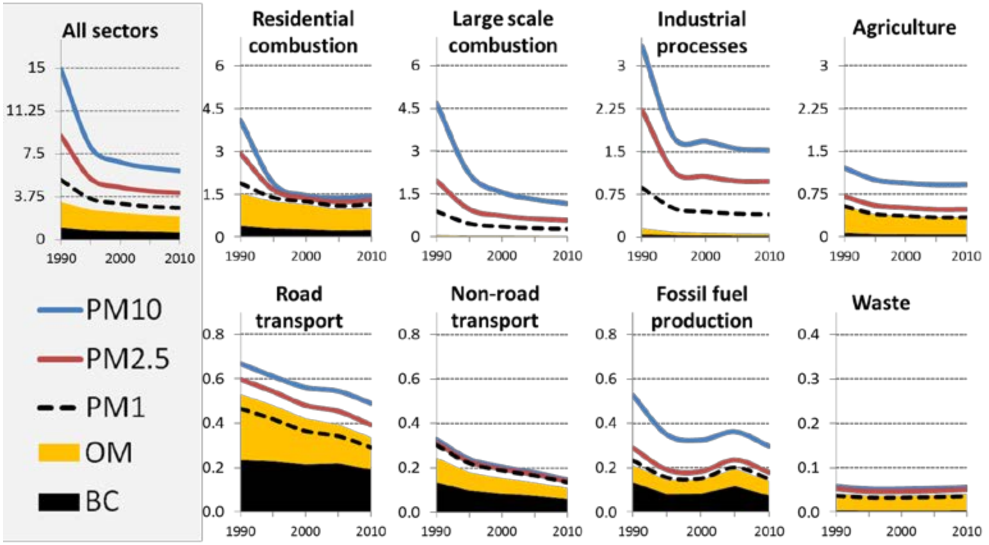


**Figure S6.6.** Emissions of PM species by key sectors in *Europe and Russia* in the period 1990-2010 [Tg year$^{-1}$]; ECLIPSE *V5a*

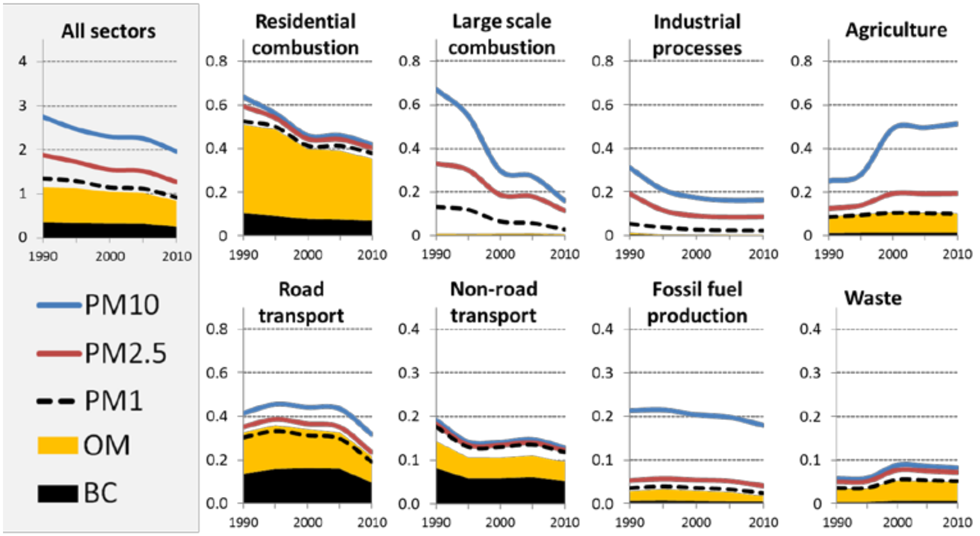


**Figure S6.7.** Emissions of PM species by key sectors in *North America* in the period 1990-2010 [Tg year$^{-1}$]; ECLIPSE *V5a*

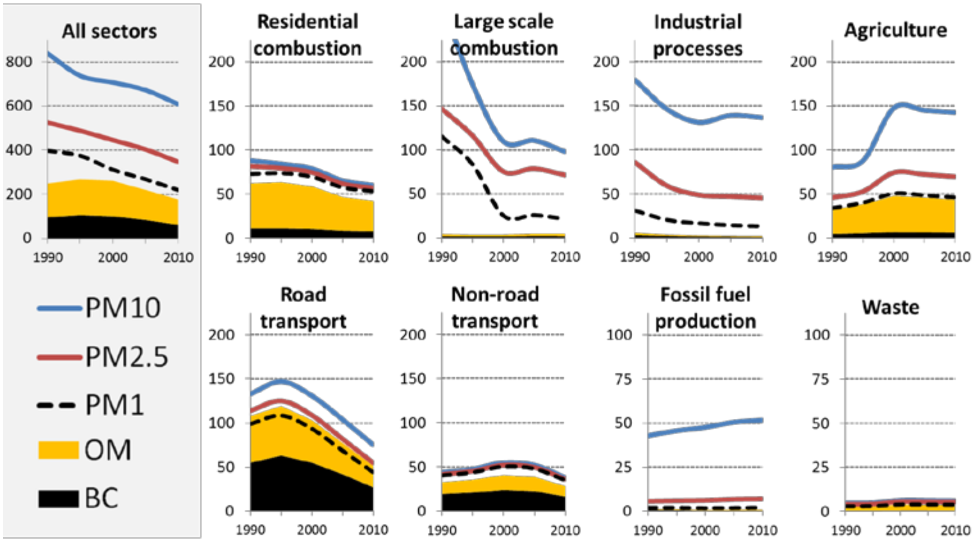


**Figure S6.8.** Emissions of PM species by key sectors in *Pacific* in the period 1990-2010 [Gg year$^{-1}$]; ECLIPSE *V5a*

## S7 Regional resolution

The spatial resolution of the GAINS model is discussed section 2.4 of the paper and the list of all 170 regions can be obtained from the online model. In principle, GAINS distinguishes single countries in Europe (exception in Russia for which European and Asian part is included separately) North America, Australia and New Zealand, for Asia several larger countries are divided into provinces or states (larger administrative units in, e.g., China, India, Indonesia, Japan, etc.) while Middle East represented as one region or (most recent versions) distinguishes Iran, Saudi Arabia, Israel, and the rest of Middle East. Africa is divided into four regions: South Africa, Egypt, North Africa, and other Africa. Latin America and Caribbean includes now 13 regions with all larger countries treated separately while Central America as well as Caribbean states are grouped in two regions. While such resolution of 170 regions is used for the calculation of emissions, the presentation of data and results differs between the on-line models available for specific world regions, e.g., for Europe and Asia the full resolution is available, while in the global model application (http://magcat.iiasa.ac.at/gains/IAM/index.login) the data and results are presented for 25 regions (Fig. S7.1). This follows closely the IMAGE model[1] resolution; often used or compatible with several global integrated assessment models.

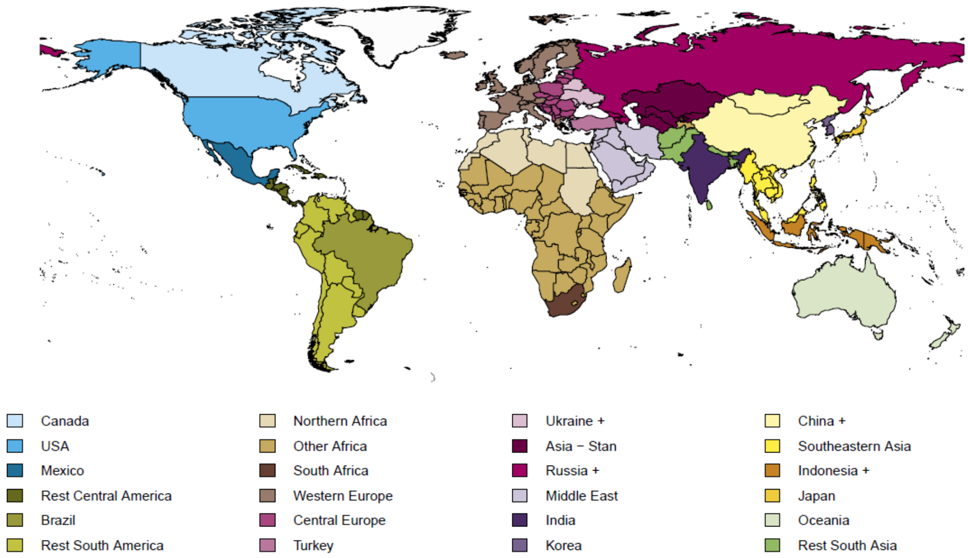


**Figure S7.1.** Regions distinguished in the global GAINS online application.

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[1] http://themasites.pbl.nl/models/image/index.php/Region_classification_map

## S8 Sectoral resolution

**Table S8.1:** Source sector resolution in the GAINS model for calculation of PM emissions

| Key source category | Source sectors | Fuel category or activity type |
|---|---|---|
| *Energy sector* | | |
| | Power plants (distinguishing small, large, old, new plants); Diesel generators; | Coal, oil, gas, biomass, waste |
| | Extraction and distribution of solid and liquid fuels (fugitive as well as combustion from gas flaring) | Coal, oil |
| | Briquette production | Production |
| *Residential combustion* | | |
| | Cooking stoves; Heating (distinguishing fireplaces, stoves, house boilers, mid-size residential boilers) | Coal, fuelwood, dung, oil, gas, agricultural residues, charcoal |
| | Kerosene lighting | Kerosene |
| | Waste (trash) burning | Waste |
| *Industrial combustion* | | |
| | Iron and Steel; Pulp and Paper; Chemical; Non-ferrous metals; Non-metallic minerals (excl. Bricks); Other | Coal, oil, gas, biomass, waste |
| *Industrial processes* | | |
| | Iron and steel industry divided into: Pig iron; Coke ovens; Agglomeration plants – pellets; Agglomeration plants – sinter; Open hearth; Electric Arc; Basic oxygen; Rolling mills; Cast Iron | Production |
| | Non-ferrous metals (copper and nickel smelters); Primary aluminium; Secondary aluminium; Cement; Lime; Carbon black production; Glass production; Mineral fertilizer production; Brick manufacturing; Pulp and paper | Production |
| | Refineries | Crude oil throughput |
| | Handling and storage of bulk industrial and agricultural products (fugitive) | Million tons of products |
| *Road transport* | | |
| | Passenger cars and vans; Light duty vehicles; Heavy duty vehicles; Busses; Motorcycles (4-stroke); Mopeds (2-stroke) | Gasoline, diesel, CNG, LPG, km driven (for calculation of non-exhaust emissions) |
| *Non-road transport* | | |
| | Agricultural and forestry; Construction and mining; Railways; Inland navigation; Coastal shipping; Aviation (landing and take-off); 2-stroke engines (e.g., in household, forestry, etc.); Other land based machinery | Diesel, gasoline, CNG, jet fuel and kerosene, heavy fuel oil, coal |
| *Agriculture* | | |
| | Arable land operations | Arable land area |
| | Livestock housing | Cattle, pigs, poultry |
| | Open burning of agricultural waste | Waste burned |
| *Other* | | |
| | Fireworks; Cigarette smoking; Barbeques; Cremation | Population |
| | Construction (fugitive) | Constructed area |

## S8 Comparison of regional estimates with selected studies

The table S8.1 provides ECLIPSE *V5a* PM estimates for selected regions and years (from the period 2000-2010) and compares them with selected regional peer-reviewed studies.

**Table S8.1:** Comparison of regional estimates for anthropogenic [a] emissions of PM species, Gg year$^{-1}$

| Region – (Source) – Year | PM10 | PM2.5 | PM1 | BC | OC |
|---|---|---|---|---|---|
| Global | | | | | |
| *This study – 1995* | 57830 | 43762 | 35902 | 6206 | 11949 |
| (Bond et al., 2004) - 1996 | | | | 4997 | 10481 |
| *This study - 2000* | 58366 | 44613 | 36741 | 6595 | 12449 |
| (Bond et al., 2013) - 2000 | | | | 4870 | |
| *This study - 2010* | 62537 | 47843 | 37819 | 7264 | 13548 |
| HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010 | 50292 | 32761 | | 5525 | 13581 |
| China | | | | | |
| *This study - 2000* | 18061 | 13554 | 11685 | 1646 | 3487 |
| (Cao et al., 2006) - 2000 | | | | 1496 | 4211 |
| (Streets et al., 2003) - 2000 | | | | 1049 | 3385 |
| (Klimont et al., 2009) - 2000 | | | | 1345 | 3205 |
| (Lu et al., 2011) - 2000 | | | | 1244 | 2823 |
| (Ohara et al., 2007) - 2000 | | | | 1093 | 2563 |
| (Bond et al., 2013) - 2000 | | | | 1200 [b] | 2800 [b] |
| (Zhang et al., 2006) - 2001 | 17120 | 12100 | | | |
| *This study - 2005* | 21087 | 15593 | 12428 | 1813 | 3552 |
| (Zhang et al., 2009) - 2006 | 18223 | 13266 | | 1811 | 3217 |
| (Klimont et al., 2009) - 2005 | | | | 1366 | 2812 |
| *This study - 2010* | 21827 | 16019 | 11564 | 1915 | 3589 |
| (Lu et al., 2011) - 2010 | | | | 1838 | 3907 |
| (Kurokawa et al., 2013) - 2008 | 21606 | 14514 | | 1589 | 3081 |
| (Guan et al., 2014) - 2010 | | 12100 | | | |
| HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010 | 16615 | 12199 | | 1764 | 3384 |
| (Kondo et al., 2011) - 2008 | | | | 1940 | |
| India | | | | | |
| *This study~~,~~ - 2000* | 8654 | 6472 | 5016 | 884 | 1596 |
| (Streets et al., 2003) - 2000 | | | | 600 | 2837 |
| (Ohara et al., 2007) - 2000 | | | | 795 | 3268 |

| Region – (Source) – Year | PM10 | PM2.5 | PM1 | BC | OC |
|---|---|---|---|---|---|
| (Klimont et al., 2009) - 2000 | | | | 842 | 1887 |
| (Lu et al., 2011) - 2000 | | | | 736 | 1990 |
| (Bond et al., 2013) - /2000 | | | | 500 [b] | 1600 [b] |
| (Reddy and Venkataraman, 2002a, 2002b) - 1998-99 | | 4300 | | 380 | 1250 |
| *This study~~,~~ - 2005* | 7952 | 5957 | 4700 | 908 | 1630 |
| (Zhang et al., 2009) - 2006 | 4002 | 3111 | | 344 | 888 |
| (Klimont et al., 2009) - 2005 | | | | 1029 | 2132 |
| *This study~~,~~ - 2010* | 8061 | 6032 | 5091 | 1022 | 1755 |
| (Lu et al., 2011) - 2010 | | | | 996 | 2582 |
| HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010 | 8280 | 6230 | | 1019 | 2530 |
| (Kurokawa et al., 2013) - 2008 | 6651 | 4884 | | 713 | 2286 |
| Europe [c] | | | | | |
| *This study - 1995* | 6905 | 4584 | 3071 | 675 | 1021 |
| (Kupiainen and Klimont, 2007) - 1995 | | | | 717 | 1053 |
| (Schaap et al., 2004) - 1995 | | | | 760 | |
| (Bond et al., 2004) - 1996 | | | | 678 | 947 |
| *This study - 2000* | 5579 | 3843 | 2668 | 618 | 910 |
| (Kupiainen and Klimont, 2007) - 2000 | | | | 680 | 996 |
| (Kupiainen and Klimont, 2004) - 2000 | | | 2772 | 672 | 988 |
| *This study - 2010* | 5008 | 3471 | 2393 | 562 | 806 |
| TNO-MACCII (Kuenen et al., 2014) - 2009 | 4694 | 3199 | | 548 | 906 |
| HTAP_v2 (Janssens-Maenhout et al., 2015) [d] - 2010 | 2951 | 2133 | | 382 | 638 |
| LRTAP reporting (www.ceip.at) - 2010 | 4784 | 3250 | | | |
| Russian Federation | | | | | |
| *This study - 2010* | 2108 | 1368 | 815 | 170 | 213 |
| HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010 | 562 | 313 | | 60 | 42 |
| (Huang et al., 2015) - 2010 | | | | 224 | |
| Russian Federation – European part only | | | | | |
| *This study - 2010* | 1090 | 734 | 427 | 71 | 122 |
| LRTAP reporting (www.ceip.at) - 2010 | 569 | 367 | | | |
| US | | | | | |
| *This study - 2000* | 1954 | 1296 | 949 | 289 | 388 |
| (Battye et al., 2002) - 1999 | | | | 430 | |
| (Reff et al., 2009) - 2000 | | | | 440 | 960 |
| (Bond et al., 2013) - 2000 | | | | 350 [b] | 500 [b] |

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| Region – (Source) – Year | PM10 | PM2.5 | PM1 | BC | OC |
|---|---|---|---|---|---|
| *This study - 2010* | 1630 | 1027 | 727 | 201 | 308 |
| HTAP_v2 (Janssens-Maenhout et al., 2015) - 2010 | 1973 | 1640 |  | 295 | 471 |
| (US EPA, 2016) [e] - 2011 | 2847 | 1909 |  |  |  |
| (US EPA, 2016) [e] - 2014 | 2830 | 1875 |  | 280 | 602 |

[a] Based on the information available in the quoted studies, all presented estimates exclude forest fires but include agricultural burning, unless stated otherwise; [b] Excluding agricultural burning; [c] Includes European part of Russian Federation (except HTAP_v2); [d] Excluding any territories of Russian Federation; [e] ~~Including wildfires and prescribed burning; [f]~~ Excluding wildfires and prescribed burning, unpaved roads, and construction dust

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