

Interactive comment on “Global anthropogenic emissions of particulate matter including black carbon” by Zbigniew Klimont et al.

Zbigniew Klimont et al.

klimont@iiasa.ac.at

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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

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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 tra –diesel, road tra – 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

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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.

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; simi-

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larly in some provinces (states) in India, Pakistan, or Nepal. The data for that originates from GAINS-Asia related studies as well as the recent work 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. 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. 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.

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

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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.

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

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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.

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

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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 CO₂ (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

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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 some 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 PM_{2.5} 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 clarify 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."

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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 to differentiate 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

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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

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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

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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,

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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.

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