The authors' response to the comments by Referee 2

The referee's comments are presented in *italic* font, followed with our responses

We thank the referee for a thorough and constructive review, which clearly improves our manuscript. Below we reply to the comments by the referee and describe how they are captured in the manuscript.

The authors add primary aerosol particle number distributions to an emissions scenario model (GAINS). Number emissions are presented for a group of components (e.g. sulphate, OC) and explicitly for black carbon (BC). The primary emissions of number are based on a three-mode representation of the aerosol smaller than 1 um diameter. The mass emissions already in the model are distributed according to various observations in the literature for primary particle emissions. The focus is on the year 2010 and those estimates are compared with estimate for 2020 and 2030 based on future scenarios. Overall, I believe it is a very useful contribution to the literature and appropriate to ACP. The paper is generally well presented, but there are several places where some improvements are needed. I have several minor comments.

Specific comments:

1) Page 1, line 16 - Define "GAINS".

Done.

2) Page 2, line 12 – Avoid "Clearly" (and use of similar terms in other places). Arctic Haze is one example of a broad aerosol that has more numbers >100 nm than smaller. Biomass burning is another aerosol that provides more larger particles. Most often, UFP dominate the numbers, but not always.

We agree and have made the following changes according to this suggestion by the referee:

P2, lines 12-16: Sentence modified and divided in two sentences: "Aerosol number concentrations are typically dominated by particles in ultrafine particle (UFP) size range, with $d_p < 0.1$ µm, or the smaller end, roughly < 0.3 µm, of fine particles (FP, here 0.1 – 2.5 µm). On the contrary, the mass concentration depends mostly on the larger and heavier, but typically fewer FP, with $d_p > 0.1$ µm (see Fig. 1 for schematic representation). "

P11 line 7: deleted "clearly"

P11 lines 19-20: "clearly" taken out and the beginning of the sentences modified as: "In 2010, China had by far the major PN emissions with 40 % estimated share of the global emissions,…"

3) Page 2, lines 16-19 – In fact, correlations of particle volume (and sulphate mass concentration) and number have been published (e.g. Leaitch et al., Tellus, 1986; Hegg and Russell, JGR, 2000), and these scenarios may well exist as often as not.

We find that the articles the referee points at are not entirely relevant for this comparison. The number concentrations showing good correlations with aerosol sulphate mass in Leaitch et al. (1986) are of particles with diameters above (roughly) 170 nm, which doesn't indicate the correlation would hold for the total particle number concentration. Also Hegg and Russell (2000) have number concentration measurements only for particles with diameters larger 120 nm, and these measurements are made in marine environments.

We have added to the manuscript two references showing poor correlations between PM and PN, and reformulated the sentence in question as:

P2, lines 16-20: "Because the particles in different size ranges originate from different sources and atmospheric processes impact them differently, the particle number (PN) concentrations and particle mass concentrations (PM, e.g. $PM_{2.5}$ describing mass concentration of particles with $d_p < 2.5$ µm) are often poorly correlated even if considering only stationary measurements (e.g. Rodriguez et al., 2007; Rodrigues and Cuevas, 2007)."

4) Page 3, lines 1-3 – Explain what you mean by neglecting the warming from BC due to their ability to form cloud droplets. This is an important point, but it is poorly described here.

Neglect was an improper choice of word here. We have modified the sentence and added the citation later pointed out by the referee:

P3, lines 1-4: "…e.g. depending on the initial sizes and atmospheric growth of black carbon particles, the net-warming effect of the BC-rich particles can gradually change, either partly or entirely, and become net-cooling when the particles start to act as CCN and form cloud droplets (e.g. Chen et al., 2010)."

5) Page 3, lines 3-5 – Discussions of the impact of reducing sulphur emissions started at least as far back as 1989 (Wigley, Nature).

This reference is added (p3, line6).

6) Page 3, lines 5-8 – Please re-write this sentence so the meaning is clear.

The sentence rewritten as:

P3, lines 7-10: "However, the changes in aerosol-cloud interactions have been so far (if not ignored) assessed by assuming similar relative changes in particle mass and number emissions, which leads to incorrect results if the actual size distributions of emitted particles change."

7) Page 3, line 10 – New particle formation can result from primary emissions. Just refer to this as particle nucleation in the atmosphere.

We have responded to comment 7 below, together with the response to comment 8.

8) Page 3, Lines 12-15 – Another sentence needing clarification. I believe you intend this as an indication of particle formation during direct emissions, but it follows the sentence about nucleation in the atmosphere without connecting with direct emissions.

We don't want to use primarily the term "nucleation", because it is not necessarily correct term for the new particle formation (if the formation occurs in kinetically limited, energetically barrierless process, the term nucleation is incorrect). On the other hand, we want to express that (regional scale) atmospheric and (small scale) combustion plume new particle formation are in principle the same process. We have modified the sentences pointed out by the referee and added the explanation also to Sect. 2.2. as follows:

P3 lines 12-22: "New particle formation (i.e. nucleation) produces particles with diameters below 2 nm (0.002 μm) from vapours such as sulphuric acid, organic vapours and nitrogen containing bases. This can happen both during regional scale atmospheric new particle formation events and at a smaller scale, for example in combustion plumes, when vapours suddenly cool immediately upon their introduction to ambient air. In this work, the latter, particles formed during the initial cooling and rapid dilution after the vapours are emitted to atmosphere, are also considered primary particles in addition to those emitted directly in particle phase. Somewhat larger UFP particles, still in nucleation mode size range, are formed e.g. in new particle formation processes occurring already before they are emitted to the atmosphere and thus producing cores for cooling vapours to condense on (e.g. Rönkkö et al., 2007; Lähde et al., 2010)."

P7 lines 5-10: "The emission factors and emissions described both in TNO work and in this study include both the particles emitted to atmosphere directly in particle phase, as well as those formed from vapours immediately after the emission during the rapid cooling and dilution of the exhausts. We consider here particles of both these types as primary particles. The uncertainties related to the emission factors in terms of particles formed immediately after the emissions are discussed in Sect. 4.1."

9) Page 3, line 32 – Comment 3 above relates to this also.

Responded above for comment 3).

10) Both Amann references on pages 4 and 5 – The first refers to EMS and the second to TSAP. Both of these need explanations.

The first, referring to the EMS article, is given as a reference for the GAINS model, which should be clear from how it is positioned in the text. For the second one, TSAP, we have added explanation:

P5, lines 9-13: “The GAINS model has been to support the Commission in the review of the Thematic Strategy on Air Pollution (TSAP; European Commission, 2005) and its related legal instruments on ambient air quality and national emission ceilings through modelling of emission baselines and scenarios for different policy options and their related impacts (Amann et al., 2013).”

11) Page 6, line 26 – define TNO.

Done.

12) Page 6, line 28 to page 7, line 3 - You refer to sizes and sectors using "i", which is confusing. In equation 1, "i" is used to refer to a region, whereas in equation 2 it refers to a size class. Make it easier.

Size classes are now referred with n.

13) Page 6, Line 27 – Add a sentence or two to indicate what factors are used to distinguish primary emissions without influence from secondary processes.

We have added the following sentence:

P7 lines 5-10: “The emission factors and emissions described both in TNO work and in this study include both the particles emitted to atmosphere directly in particle phase, as well as those formed from vapours immediately after the emission during the rapid cooling and dilution of the exhausts. We consider here particles of both these types as primary particles. The uncertainties related to the emission factors in terms of particles formed immediately after the emissions are discussed in Sect. 4.1.”

14) Page 7, lines 4-6 - EFpn:S? Is it intended as a plural of EFpn?

Yes, EF_{PN}:s stands for plural of EF_{PN}.

15) Page 8, line 23 – “New PSDs were” or “A new PSD was”?

Corrected, p9 line 6; “A new PSD was…”

16) Page 9, lines 3-5 - By black carbon mode, I assume you mean "pure" black carbon mode. Please clarify in the text.

Sentence clarified:

P9 lines 17-20: “Two different size distributions were determined, one for the whole particles in black carbon mode (BC_{mode}), which considers both the black carbon cores and the condensed material on them, and one for the black carbon cores of these particles (BC_{core}). “

17) Page 9, lines 9-14 - The Sorensen et al. work focussed on the morphology of larger particles. Their techniques and interest limited the discussion of smaller particles, and *indeed their micrographs indicate BC particles smaller than 50 nm. See Liggio et al. (Environ. Sci. Technol. 2012, 46, 4819−4828). It may be that your modal representation includes sufficient BC at sizes smaller than 50 nm, but the discussion needs to be a little more objective on this point.*

It is correct that our choice for minimum GMD for BC modes is a rough estimate and may exclude some BC-related modes from some sources, e.g. gasoline vehicles. We inspected the main sources with GMDs below but close to 50 nm and the results would not be much different if the limiting GMD was set e.g. to 40 nm. However, this does not mean that our approach is perfect, but we find it is something we can improve in the next version of emission factors. Accordingly, we added the reference cited by the referee and modified the text as:

P9 lines 24-32: "Of the combustion sources, only the modes with geometric mean diameters (GMD) equal to or above 50 nm were taken as black carbon modes. This rough estimate for a minimum GMD was chosen, because the agglomeration in BC formation produces a roughly lognormal mode and we assumed that would not form particles in the smallest size ranges of the modes with GMD below 50 nm (Sorensen et al., 1996; Kholghy et al., 2013). This assumption seems reasonable for diesel-fuelled vehicles, but might not be valid for gasoline-fuelled vehicles (Liggio et al., 2012). However, as the global emissions from diesel-fuelled vehicles are found to dominate the transport emissions, we will leave the further improvements on defining the black carbon modes to future studies."

We also added the following sentences to the end of Sect. 3.4.1.:

P17 line 30 - P18 line2: "It is to be noted that the method of defining the source-specific BC modes was approximate, as discussed in Sect. 2.3, and some of the sub-50 nm particles here defined as non-BC particles might in reality have a BC core. Even though this possible underestimation of smaller BC particles is unlikely to concern the diesel emissions (Liggio et al., 2012), which is the main source for BC number emissions, the black carbon size distributions from other sources should be assessed in more detail in future."

18) Page 9, lines 18-21 – There has been significant debate as to what constitutes primary OC. This should be reflected in the response to comment 13 above.

In these estimates we made the rough assumption that all OC in the studied size range is formed through condensation during the initial cooling of the exhaust gases. In reality, there is also a small fraction of solid PM_{OC}. For example, organic particles can also be emitted as solid particles if the combustion conditions are poor. This solid PM_{OC} is always part of the primary OC independent of what the measurement method defines on cooling or dilution. It is however, a relatively small fraction of the total PM_{OC} if some dilution and cooling are taken into account. For example, for conventional woodstoves, one of the most important categories in Europe, the average solid particle emission factor is 150 g GJ^{-1} (range 49–650) – this includes a small fraction solid PM_{OC} – whilst the average of the dilution tunnel measurements, that include both solid and condensable particles, is 800 g GJ^{-1} (range 290–1932). (Denier van der Gon et al., 2015, Nussbaumer et al., 2008) The gap between the 150 and the 800 g GJ^{-1} is entirely due to condensable, non-solid OC showing that this is by far the dominant PM_{OC} fraction.

19) Page 9, line 30 – "except for BC formed through condensation". This is confusing. I assume you mean PM1 minus the BC component. Please clarify.

Modified as:

P10, line 16: "for the case of all PM_1, except for BC, assumed to be formed through condensation"

20) Page 18, lines 23-24 – "are smaller"

Modified: P19 lines 23-25: "In Europe and Northern America, the overall uncertainties, even though significant in absolute values, are smaller in comparison to the other continents, both in terms of current and future emissions."

21) Page 19, line 19 – ". . . the primary emissions of . . ."

Done (p20, line 21).

22) Page 21, line 17 – "primary emissions"

Done (p22, line 25).

23) Page 21, line 21 – what do you mean by aerosol formation: secondary processes; nucleation?

We mean both vapours condensing on the pre-existing particles and nucleation (referred to as new particle formation, see answer to comments 7 and 8). Clarified in the text:

P22, line 30 – P23, line 2: "…in the areas of low anthropogenic primary emissions the natural emissions and atmospheric aerosol formation (both in terms of vapours condensing on pre-existing particles and formation of new particles from vapours) play a relatively more important role…"

24) Page 22, lines 1-15 – I am surprised that you have chosen to focus only on the warming aspect of BC, when your work seems suited to address the question about how much BC affects the number distributions and therefore the number concentrations of CCN; see Chen et al 2010, GRL. Estimating how the total number concentrations compare with the BC number concentrations from Figures 6 and 7 is very difficult, and in Figures 8 and 9 you only refer to BC mass. I would like to see a plot comparing the average number distribution from Figure 6 and the average BC mode and core number distributions from Figure 7. Of course there are still particles formed from secondary processes to be considered, but more knowledge of the importance of BC to primary emissions would be immensely useful.

We did not intend to discuss only the warming aspect of BC, the changes in FP (roughly CCN) number emissions was discussed in the same sentence. However, the connection between BC mass and FP number emission was not pointed out. Thus, we have added the following:

P23, lines 18-20: "The predicted changes in BC mass emissions and PN emissions suggest that, even though the BC particles can act as CCN after atmospheric aging (Chen et al., 2010), the overall global decrease in BC mass emissions does not lead to similar decrease in number emission of FP."

It is true that the comparison between BC and non-BC particles was not easy enough from the original figures. Thus, we have substituted figure 7c with the figure the reviewer asked for, i.e. PSD of total global PN emissions and total global number emissions particles containing BC core. We found that the original Fig. 7c was so similar to 7b that the existing explanation of the difference was enough to describe it (P17, lines 24-29: "The difference between the assumptions of the composition of the coating of BC cores, i.e. the choice between coating including only OC and coating including all PM_1 except BC, was significant only in industrial combustion emissions, for which the BC core mode shifted to much smaller sizes (from ~100 nm to 30-40 nm) when assuming all PM_1 is condensed on BC cores.").

References:

Denier van der Gon, H. A. C., Bergström, R., Fountoukis, C., Johansson, C., Pandis, S. N., Simpson, D., and Visschedijk, A. J. H.: Particulate emissions from residential wood combustion in Europe – revised estimates and an evaluation, Atmos. Chem. Phys., 15, 6503-6519, doi:10.5194/acp-15-6503-2015, 2015.

Nussbaumer, T., Czasch, C., Klippel, N., Johansson, L., and Tullin, C.: Particulate Emissions from Biomass Combustion in IEA Countries, Survey on Measurements and Emission Factors, International Energy Agency (IEA) Bioenergy Task 32, Zurich, 2008