

The authors' response to the comments by Referee 1

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 present an implementation of global particle number emission factors to the GAINS model. Such work is very important for atmospheric aerosol studies, to move more towards a complete description of the aerosol also from the emission side. The manuscript gives an overview of the applied methodology, and also gives a general description of the results of the application. The main sources of uncertainty are also discussed in some detail. The manuscript is an valuable addition especially for modellers in the atmospheric community, and it should be published. There are some comments that should be answered before publication, though.

Generally, the manuscript (according to the title) discusses primary particles. As also discussed in parts in the text, some sources emit vapours that in some conditions quickly form particles. For some sources, this is not captured by the emission factors, because such particles are purposefully removed e.g. by heating, while for some sources these particles will be present. This is an issue for example for the sulphur-related emissions. Is there a distinction between the "actual-primary" and "quickly-formed secondary" particles done in the current work? Are there possible errors in the emission estimates due to this?

In this work we apply reported particle number size distribution measurements within fresh plume, instead of stack measurements, for deriving the most part of the emission factors. Thus, the quickly formed secondary particles are included in the emissions factors, as far as the measurement methods have allowed their detection. However, with this basis for emission factors, we are not able to directly make distinction between the "actual-primary" and "quickly-formed secondary" particles as suggested by the referee (clarified in the revised manuscript on page 3, lines 16-18, and page 7, lines 5-10). Our calculations of black carbon containing particles would, in principle, allow a rough kind of distinction towards what the referee suggests, if all the particles smaller than the assumed black carbon mode were taken as "quickly-formed secondary" particles. We decided not to do this in this manuscript, partly due to the other remark made by the referee: there are differences in the measurement techniques (in terms of how and if the non-solid particles are detected) used in the literature applied for the determination of the GAINS emission factors. We find that the deeper analysis of the related uncertainties and the revision of the emission factors with respect to the emissions of these non-solid, e.g. sulphur-related, particles requires further work and we highlight the importance of pursuing this in the near future.

The possible errors arising from partial inconsistency treatment of quickly formed

secondary particles between different source sectors are discussed in Sect. 4 under title “Effect of sulphur on PSDs and emission factors”. By lifting these uncertainties as the second of the three general sources of uncertainties in Sect. 4.1, we point out the importance of this issue and hope it will allow for future funding on this topic. We modified Sect. 4 in terms of dividing the sources of uncertainties under topics of “General” and “Sector-specific” causes for uncertainties, Sub-sections 4.1 and 4.2, respectively (pages 20-22). With this, we moved the paragraph “Effects of ambient conditions on emissions” to the end of sub-section 4.1 (more on the changes in this paragraph, please see the answer for the specific question below).

Point-by point comments:

p4, line 3: the authors refer a couple of times to coagulation as the main sink of particle number. The study that they are referring to (Kerminen et al., 2001) refers to recently-born particles, and should not be generalized to all sub-100 nm particles. For example, particles with a diameter of 50 nm have ca. 50-fold lifetime due to coagulation when compared to 5-nm particles, and other loss mechanisms will become important as particles grow. Please change the text to reflect this. Coagulation will of course be important immediately after emission and at high concentrations, and the effect of increasing number with decreasing mass emission is thus possible.

We have made the following (underlined) additions to the text according to this comment:

P4, line 8: “Additionally, because the main removal mechanism of the smallest of UFP in the atmosphere is their coagulation to larger particles (e.g. Kerminen et al., 2001),...”

P23, lines 3-4: “...the most efficient sink for the smallest of aerosol particles in nucleation mode is their coagulation with larger particles...”

p 7 line 1: it would be nice to know the number and spacing of the size sections, especially also the lower limit of the size sections.

The information on the size sections was unintentionally left out from the manuscript. We have added a table (Table 1) showing the size ranges of the size classes and made the following additions to the text:

P7, line 16-17: “The diameter ranges for the size classes applied for the GAINS emissions are shown in Table 1.”

P8, line 7: “(see Table 1)”

P7 l8: uncertain language: this statement seems to me a bit vague. Measurement methods are generally less uncertain in these size ranges. As explained later, authors mean that even a small error in the number of near- 1um particles causes a large error in the number of smaller particles when converting mass concentrations. This is a valid and

good point, but the wording here should be fixed.

The indicated sentence has been removed, and the following sentence has been modified as:

P7, line 22-27: “However, deriving an EF_{PN} directly from the EF_{PM1} would make the EF_{PN} very sensitive to the estimated number of close to 1 μm particles, since their mass is significantly larger in comparison to the mass of those with diameter below or around 100 nm. Thus, emission factors for PM in the size range 10-300nm ($EF_{PM0.3}$) were first derived from EF_{PM1} based on literature on emission mass size distributions and particle densities (M. Kulmala et al., 2011, H. Denier van der Gon et al., 2010).”

P7 l18 Which TNO study is meant? Two references were originally given.

“Study” is now replaced with plural and references are repeated (p7, line 31).

p12 l9-13: What is meant here? Surely the emissions are dependent on the sulphur removal techniques as well as the fuel content, but this has not been accounted for in the modelling? Or is something else meant here? These sentences should be clarified, as well as the the reasoning for this factor only causing an underestimation.

We meant that the applied emission factors are not dependent on fuel sulphur content or sulphur removal technologies, but the choosing of the words was wrong. This is now corrected (p12, line 24: “the developed emission factors for (coal-fired) power plants are not dependent...”).

The emission factors for power plant emissions are based on the previous TNO studies, which have focused on European emissions. Since in Europe the coal-fired power plants are in practise all equipped with some level of sulphur removal technology, the sulphur related PN emissions are estimated to be minor. However, in countries where this is (partly) not the case, the sulphur related addition to power plant emissions can be expected to be significant or even dominant. This has now been clarified in the manuscript:

p12, lines 26-30: “The applied power plant emission factors are designed for power plants in Europe, where sulphur removal technologies are in place. This may cause significant underestimation in the emission estimates for power plants using high sulphur fuels (for other power production sources than coke production) in many parts of the world, where a significant fraction of the power plants are not equipped with such technologies.”

p13 l4: please also give a pointer to the specific reason for the inconsistency discussed in Sect 4.

We are not sure which inconsistency the referee is pointing at, since the given line (p13 line 4 in the original manuscript) is the title of the section “Industrial combustion and processes”, which ends with a pointer to Sect. 4.

However, it seems appropriate (and the referee might have meant) to add a pointer to the uncertainties related to the connection between sulphur emissions and PN emission factors in the end of the Sect 3.2.1 discussed above. We replaced the last sentence of Sect 3.2.1. with:

P13, lines 3-5: “These uncertainties, also influenced with the general uncertainties related to the representativeness of the PN emission factors for nucleation mode sulphate/sulphuric acid particles, are discussed in more detail in Sect. 4.”

p14 line 31: neglected is probably not the right word here?

No, it is not. Replaced with “invalidated” (p15, line19).

page 15, section 3.3. and Fig 5: I think using the measure of particles/grid cell is not the right choice, especially if this is then used for comparisons between different geographical areas. This because the grid area is different near the equator than near the poles (the gridding is based on latitudes and longitudes), and for comparison purposes the emissions should be normalized by the grid cell area.

This is right. We have replotted the figure in units $\#/km^2$. With this we noted that there was a mistake in text saying that the emissions ranged in span from 10^{19} to 10^{25} per grid cell. This span was not the original, but the colour values for the figure were set to this (i.e. all values below 10^{19} were shown as 10^{19}). We have reviewed this sentence as follows:

P16, lines 16-18: “The gridded emissions ranged in a span of various orders of magnitude (note the logarithmic colour axis in Fig. 5, where the values below 10^{16} particles/ km^2 /year are shown as having the value of 10^{16}).”

p18, l23: "In Europe and Northern America, the overall uncertainties can be estimated to be relatively small, both in terms of current and future emissions." This is a quite optimistic sentence. The word 'relatively' makes it probably correct in terms of comparisons with other estimates, but there remain several factors in measurement technologies, changes in abatement technologies, ultrafine particle concentration variability etc. that I think that a statement that uncertainties are small is yet preliminary.

Correct. We have modified the sentence pointed out by the referee as (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.”

p19 l10 -> "This may result ..." this is an interesting point. How does the GAINS model see particles that are formed due to a decrease CS from the original source? Such particles could also be considered secondary particles, should such particles be covered by emission inventories? See also general comments.

For the PM-based emission factors the change in condensation sink CS (from the same source) due to technological improvements doesn't affect the emissions at all, but the relative change in each size bin is directly proportional to the change in PM (mass) emission factor. For direct emission factors see the answer for the general comment.

p20 line 6, "Effects of ambient conditions on emissions": This is an important point. Here, the authors consider only a very limited temperature effect on the emissions; actually, also many other factors, such as the existing aerosol concentration, might affect the emission, especially if some secondary particles are also considered in the emission factors. This could be elaborated on in this section.

We have underlined that the explained temperature effect is an example (P20, line 2), and added the following to the end of this section:

P21, lines 13-18: "Also the particle concentrations prior to emission can be presumed to affect the PN number emissions (at least when the immediate formation of secondary particles are considered as PN emission), due to the competition of (emitted) vapour uptake between new particle formation and condensation to pre-existing particles. These kinds of effects are, however, issues for future research and their impact cannot be implemented directly to the GAINS model."

p22 l32: remove "the transport emissions". Also, remove hyphen from "Southern America"

Done.

Table 1, l4 (page 33): This sentence is very complex: what is decreasing or increasing, and what causes what? How can high FSC cause effects when it is assumed to be replaced by low FSC? This is explained better in the text, but understanding the table is hard with this caption.

This explanation has turned out to be extremely difficult to be put in words. We have now rewritten the caption as follows:

P35, lines 4-11: "The relative change in annual road transport PN emissions in comparison to the CLE scenario, if (in addition to the technological advancements described in CLE scenario) all the diesel fuel (consumed in road transport) is assumed to be replaced with ultra low sulphur content –fuel (FSC = 10 ppm). The lowest row shows the change in total emissions from all sources. Note that e.g. in Europe, the impact increases with time, because in the CLE scenario the emissions decrease drastically in most countries, but a small share of high FSC fuel remains present in some (non-EU) countries. Thus, the proportion of the high FSC contribution to total emissions in the CLE scenario increases with time."