Referee comment to "A global analysis of climate-relevant aerosol properties retrieved from the network of GAW near-surface observatories" by Laj et al.

Response to Anonymous Referee #1

We would like to thank Anonymous referee #1 for the very useful comments on the manuscript. You will find below our specific answers to the different points raised in the review. All modifications are noted in red in the new version of the manuscript, sent to AMT.

The results of a large effort to summarize the climate-relevant in situ aerosol properties available at all sites connected to the GAW network are presented. The growth of the global base over the past few decades is shown as well as the increase in the usage of the data over the past ten years. Values of absorption, scattering, and SSA are compared for the wide range of existing sites. Seasonal data and annual trends are also presented. All in all, it is a substantial effort that is definitely worthy of publication in AMT. Relatively minor comments are listed below.

Table 1. Should the description of CCN say "CCN number concentration is sometimes approximated using the fraction of particles larger than a given diameter from the particle number size distribution AND AN ASSUMED CHEMICAL COMPOSITION"?
In principle, this is correct. However, it is true that many studies are approximating the CCN number concentration using the fraction of particles larger than 100 nm, without any consideration on their chemical composition/hygroscopicity. This is often the case for long-term records where chemical composition is not necessarily avaialable with the required time resolution (1h). We have made this point clearer in the Table adding « neglecting the influence of particle chemical composition »

Line 225: ". . .but also including sites IN other WMO regions. . ."
This is corrected in the manuscript

Line 249: "..many of them ARE no longer documented. . ."
This is corrected in the manuscript

Line 271: Analyzes instead of analyses?
In the paper, we have constantly used the UK wording with « s » and not the US with « z ». So we did not implement that change.

Line 271: Define SARGAN here where it is first used.
This is corrected in the manuscript

Lines 320 – 325: It would be handy to have a reference to the relevant report of the GAW measurement guidelines included in Table 2 , i.e. WMO/GAW Report #200 for light scattering and absorption.
References to all WMO/GAW reports are actually listed in the original version of the manuscript.

Line 364: define the ultrafine and fine ranges in terms of diameter.
Ranges are now defined (10-100 nm and 100-1000 nm)

Lines 400 – 413: When was this change made, i.e. only removing data affected by instrument issues or contamination? Were older data sets amended?
The change in quality control approach was induced by the shifting objectives of WMO and thus GAW. These become visible in the shift between the GAW Strategic Plan: 2008 – 2015 and the GAW Implementation Plan 2016 – 2023. While the 2008-2015 Strategic Plan talks about "Changes in the weather and climate related to human influence" and "Risk reduction of air pollution on human health

and issues involving long-range transport and deposition of air pollution", the 2016 – 2023 GAW Implementation Plan mentions "Strengthen capabilities to predict climate, weather and air quality". This reflects GAWs shift from observations relevant for climate and long-range transport towards services and further application areas such as air quality prediction. The shift from single purpose to multi purpose quality control was thus introduced in 2016. Due to the lack of recent guidelines for GAW aerosol data quality control, this change was communicated through project meetings and bilateral contact with data providers. Due to the scientific independence of GAW stations, implementation of the new data QC approach varies. Only few data providers re-processed older data due to work load restrictions, but most have adopted the new QC policy.
To reflect this information, the text is changed to "As of 2016, it was acknowledged that atmospheric composition data serves multiple purposes and applications. This is reflected by the recommendation to only remove data affected by instrument issues or contamination during quality control, and indicate local or regional influence with a flag that leaves the data valid. This implies, for any application of WDCA data, filtering the data according to purpose is the first step. When using WDCA data, this shift in quality control approach, which may vary among stations due to their scientific independence, needs to be taken into account. Due to resource limitations, data before 2016 was mostly not reprocessed."

Lines 439 – 440: Are there references for the "manual-expansion type" and "automated version" that can be provided so the reader knows what these particle counters are?
Proper reference has been added in the new version of the manuscript : Hogan, A.W., and Gardner, G. (1968) A nucleus counter of increased sensitivity. J. Rech. Atmos. 3:59-61.

Line 484: define kerbside.
This is now defined : (near-road)

Line 540: A couple of sentences with references on different methods that have been used to calculate trends would be helpful here.
The following sentences were added to the section : « The non-parametric seasonal Mann-Kendall (MK) statistical test associated with several prewhitening methods and with the Sen's slope was used as main trend analysis method (Collaud Coen et al., 2020 submitted). Comparisons with General Least Mean Square associated with Autoregressive Bootstrap (GLS/ARB) and with standard Least Mean Square analysis (LMS) (Asmi et al., 2013, Collaud Coen et al., 2013) enabled confirmation of the detected MK statistically significant trends and the assessment of advantages and limitations of each method »

Line 551: ". . ..too high, however, OBSERVATIONS INDICATE it is. . .."
This is now corrected in the new version of the manuscript

Lines 551 – 552: What exactly is meant by "While the bias values are robust at the sites investigated..."? The bias values (i.e. model – measurement differences) are well characterized or low?
The reviewer is right, the sentence was not very clear. We have rephrased the whole sentence and the sentence before and it reads now: " This would indicate that the overall OA/OC ratio in the models is too high, although many model assume for primary OC emissions a low OA/OC factor of 1.4. Secondary organic aerosol formation increases this ratio in global aerosol burdens. Note that the biases established are for the relatively few remote sites investigated. It is currently difficult to assess, if there is a robust global bias in OA, OC or its ratio for the models in question."

Lines 565 – 569: How did the measured and modelled number size distributions compare?
We have modified the sentence to : Kok et al. (2017), showing that dust found in the atmosphere is substantially coarser than represented in current global climate models, suggest that AeroCom models do not have a sufficient coarse dust component, which suggests that dust may even have a warming direct radiative effect.

Line 570: ". . .is the evaluation OF MEASURED AND MODELLED cloud condensation C2 nuclei."
This is now corrected in the new version of the manuscript

Line 580: Provide the link to GAWSIS here where it is first mentioned.
This is now added to the manuscript

Line 587 – 589: What is the connection between not all GAW stations being able to measure all variables listed in Table 1 and SARGAN being a subset of stations in GAW? Please clarify.
This is now clarified in line 592 : SARGAN is, therefore, a subset of stations in GAW providing in-situ aerosol variables from ground-based stations.

Line 628: Where are WMO regions I, II, III, and IV? I don't think this is stated previously.
This is now provided in the text

Line 634: What is meant by "a station footprint that is large"? Is this related to its representativeness of a region? Or land type?
We have now added : (influenced by air masses transported more than 100 km away) in the text to provide a better definition of a large footprint

Line 735: ". . .for 29 of these sites IT was possible. . ."
This is now corrected

Lines 744 – 746: Is it possible to cite a reference for this assumption?
AAE=1 was chosen for the harmonization between different devices and wavelengths as suggested by Zanatta et al., (2016, https://doi.org/10.1016/j.atmosenv.2016.09.035), now added as a reference to this assumption

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Line 772: It is more commonly thought that Cape Grim is a coastal Southern site, than a Pacific site.
This is corrected in the new version

Table 3: Abbreviations shown in the plots (DE, RB, U) should be defined in the caption.
This is now defined in Caption of Figure 6

Figure 8a: The title says monthly means while the caption says seasonal means. Given the number of points, I assume it is the former.
Figure 8 has now been changed, responding to the request by Reviewer #2. The comment does not apply anymore

Figure 8: Coloring the points by month may provide useful information on under- and overestimates by the models. Also – 8a shows absorption and 8b shows scattering but scattering is introduced in the text first.
Figure 8 has now been changed, responding to the request by Reviewer #2. The comment does not apply anymore

Figure 9 caption: Provide a reference for the Mann-Kendall trend method and describe the Sen's slope estimator.
The reference to Collaud Coen et al., submitted was added to the caption and to the reference list

Lines 868 – 870: This sentence is confusing. What does "almost all stations have either statistically significant decreasing or not statistically significant trends in the absorption coefficient" mean? Does it mean that the only ss trends in absorption are decreasing trends?
Yes, the only statistically significant trends in absorption are decreasing trends. In fact, some corrections were done for the revised manuscript (companion paper Collaud Coen et al., ACP 2020, submitted) so that no station have an annual statistically significant increasing trend in absorption any longer. The sentence is the present manuscript was modified to:" The trends of the absorption coefficient are ss decreasing or not ss for all the stations."

Lines 873 – 879: It is a little frustrating that hints of interesting trends are mentioned ("Polar stations exhibit a mix of increasing and decreasing trends") without more detailed explanation. Why is scattering at ZEP, PAL, and SPO increasing on an annual basis but decreasing at BRW? If this is discussed in more detail in the companion papers that should be explicitly stated here so that the reader knows where to find further information. Also – it's not clear what an annual average represents since there may be a decreasing trend in one season and an increasing trend in another. I am thinking of sulfate in the Arctic where it is decreasing in winter/spring due to air quality regulations but could be increasing in summer due to decreasing sea ice.
The companion paper does not solve the reasons why, contrarily to ZEP, PAL and SPO, BRW has a decreasing trend in scattering coefficient. It has to be noted that ALT also has a decreasing trend in scattering. Potential reasons are however described in the paper: " PAL, the northernmost station, has a ss positive trend. PAL is geographically situated in Europe but it can be climatologically considered an arctic station (Schmeisser et al. 2018). PAL (slope=0.06 Mm-1/y) has a similar trend as ZEP (slope=0.05 Mm.-1./y), the nearest Arctic station, with the largest ss trend in summer (JJA) when PAL is largely influenced by Arctic air masses. The increasing trend at PAL may be due to increasing biogenic secondary organic aerosol formation related to emissions from the surrounding boreal forest (Lihavainen et al., 2015a), changes in circulation patterns or a larger influence of open water with increasing concentration of sea salt aerosol."

Lines 888 – 890: "...simulated trends are in agreement with SARGAN derived trends suggesting significant decreases found over North America and Europe. . ." This sounds like models are being used to validate measurements.
The sentence was changed to : For both variables, simulated trends can reproduce SARGAN derived trends suggesting significant decreases found over North America and Europe, although it must be considered that the number of models providing trends in σ_{ap} and σ_{sp} remains limited.

Lines 892 – 894: Is this supposed to say that ". . .NO statistically significant AOD and sulfate trends. . .".
It is actually **non** statistically. This is now corrected.

General comment: There is heavy use of Collaud Coen et al. (submitted) and Mortier et al. (submitted) in this paper. I am not sure of AMT's policies concerning citing results from papers that have not been published yet.
We assume AMT/ACP policy allows for referencing submitted papers. Collaud Coen et al, is now accepted for publication in ACP and it appears that companion papers Gliβ et al, and Mortier et al., are close to being accepted having to deal with mostly minor modifications.

Referee comment to "A global analysis of climate-relevant aerosol properties retrieved from the network of GAW near-surface observatories" by Laj et al.

Response to Anonymous Referee #2

We would like to thank Anonymous referee #2 for the very useful comments on the manuscript. You will find below our specific answers to the different points raised in the review. All modifications are noted in red in the new version of the manuscript, sent to AMT.

Overall comment: This paper provides the full technical descriptions and overall sum- maries of the in-situ aerosol primary datasets (total number concentration, scattering and absorption coefficients) from a global network of near-surface aerosol monitoring stations organized and maintained by the authors. The dataset itself will play a central role in evaluating the accuracy of aerosol models used for climate simulation. The dataset demonstrated for the first time the decadal decreasing trends in surface aerosol concentration over the globe. This should be important as quantitative evidence of the overall outcome of the pollution mitigation efforts in many countries performed in this period. It should be published in any case.

Major critical comment: I recommend the authors remove/shrink the presentations and discussion on the "comparison with models" (section 6.2.4, Figure 8) which may distract the reader's attention from the main story of this paper. Those results can be included in the companion papers (Glib et al., Morthier et al. submitted). The model results without descriptions of the underlying assumptions (i.e., details of parameterization, emission) are not very informative to me.

Thanks for this comment. We have actually modified and shorten the section, and also move it to a new section 7. We believe it is however important that this section is maintained in Laj et al., as both submitted manuscripts - Gliβ et al, submitted and Mortier et al., submitted - are not only using SARGAN data but other variables from other observations from the ground and from space. We consider it is important to maintain a clear Observation/Modelling section in Laj et al., so that agreements and discrepancies for the specific case of SARGAN variables can be discussed specifically. The section is now, however, a bit shorter, and graphs have been simplified to more clearly illustrate model performances to reproduce observations. It is also important to mention that some numbers were actually updated following comments from referees in the review process of Gliβ et al, and Mortier et al. Both papers are now sent back to ACP editors.

The manuscript now reads as follow :

7 Comparison with AeroCom model outputs for optical properties

The AeroCom initiative has focussed since 2002 on the evaluation of global aerosol models with observations (aerocom.met.no). The recent generation of AeroCom models has been asked to provide additional diagnostics on dry scattering and absorption coefficients at ground level. These are currently being analysed by the two companion papers of Gliβ et al. (in review, 2020) and Mortier et al. (in review, 2020) using 14 model simulations of present day (2010 emissions and meteorology) to construct an ensemble mean AeroCom model and aerosol information extracted from SARGAN surface sites. For a detailed analysis of comparison for variability and trends, readers can refer to the two companion papers. Here we simply provide an overview of the AeroCom median model ensemble used for comparison with observations for the specific SARGAN sites.

7.1 Comparison of observed and modelled optical properties of aerosol particles

Overall, the performance of the model ensemble varies greatly as a function of station location, for both scattering and absorption coefficients. Figures 8a and b compare monthly medians observations and model median ensemble results for the grid point corresponding to the station location, for scattering

and absorption coefficients, respectively. Results show a normalised mean bias (NMB) - defined as: (Model - Observation) / Observation - of, on average, -14 % between scattering by AeroCom models and observations, pointing to regional deficiencies in aerosol models. The NMB for absorption is smaller (4%) indicating a better performance for this parameter by the AeroCom models. Obviously, there is, for both scattering and absorption, a large station-to-station variability in the bias, showing either good agreement, under- or over- prediction depending on the site. There is also a significant variability of the NMB between models and observations when calculated for each season. This is also the conclusion of Gliβ et al. (in review, 2020) which quantified the biases to -34% and -20% for scattering and absorption, respectively and listed possible causes for the biases such as overestimate of scattering enhancement due to hygroscopic growth and the uncertainties in the treatment of absorption optical properties of black carbon, dust and organic aerosol. At this stage, additional investigations are needed to identify what accounts for the observed differences between model and observations.

7.2 Observed and modelled trends of aerosol optical properties

The issue of long-term trends for the aerosol in-situ optical properties is specifically addressed in Collaud Coen et al. (2020) using data from WDCA extending back to 40 years for some stations. Collaud Coen et al., (2020) derived time series of measured scattering, backscattering and absorption coefficients as well as the derived single scattering albedo, backscattering fraction, scattering and absorption Angström exponents at stations with at least 10 years of continuous observations. With respect to the previous trend assessment (Collaud Coen et al., 2013) which used data extending up to 2010, the number of stations with time series longer than 10 years has almost doubled (24 in 2010, 52 currently) so that the spatial coverage is improved and various additional environments are covered in Europe, North America and in polar regions. The few stations in Asia, Africa, South America and in Oceania/Pacific region cannot, however, be considered as representative for their continents/regions, both because of their small number and also because mountainous and coastal environments are overrepresented relative to the continental environment with rural, forest or desert footprints.

Methodologies and results are presented in detail in Collaud Coen et al. (2020) and are simply summarized here for scattering and absorption coefficients as well as single scattering albedo (Figure 9). The non-parametric seasonal Mann-Kendall statistical test associated with several prewhitening methods and with the Sen's slope was used as main trend analysis method (Collaud Coen et al., submitted). Comparisons with General Least Mean Square associated with Autoregressive Bootstrap (GLS/ARB) and with standard Least Mean Square analysis (LMS) (Asmi et al., 2013, Collaud Coen et al., 2013) enabled confirmation of the detected MK statistically significant trends and the assessment of advantages and limitations of each method. For scattering coefficient, statistically significant (ss) increasing trends are found at polar and coastal stations with rural background, pristine and forest footprints, whereas the largest statistically significant decreasing trends are primarily found at stations with mixed and urban footprints. Few mountainous stations have statistically significant scattering coefficient trends, whereas all of them have ss decreasing absorption coefficient trends. All stations have either statistically significant decreasing or not ss trends in the absorption coefficient. The single scattering albedo trends seem not to be dependent on either the environment or on the footprints, but rather on the geographic area (Collaud Coen et al., 2020).

Analysis of the long-term information provides evidence that the aerosol load has significantly decreased over the last two decades in the regions represented by the 52 stations. Currently, scattering and backscattering coefficients trends are mainly decreasing in Europe and North America and are not statistically significant in Asia. Polar stations exhibit a mix of increasing and decreasing trends. In addition to PAL, the northermost European station that can be climatologically considered as an Arctic station, ZEP and SPO also have statistically significant positive trends, whereas no statistically significant trend is found for the other Antarctic site (NMY). BRW and ALT both exhibit statistically significant negative 10-year trends. A few increasing trends are also found at some stations in North America and Australia. Absorption coefficients also exhibit mainly decreasing trends. Generally, these decreases in aerosol burden are expected to be a direct consequence of decreases in primary particles and particulate precursors such as SO_2 and NO_x due to pollution abatement policies.

The single scattering albedo is one of the most important variables determining the direct radiative impact of aerosol so that its trend analysis - derived for the first time from a large number of stations - has the largest climatic relevance. The global picture is nuanced with statistically significant positive trends

mostly in Asia and Eastern Europe and statistically significant negative trends in Western Europe and North America leading to global positive median trend of 0.02%/y. 15 stations exhibit a positive single scattering albedo trend (relatively more scattering) while 9 stations exhibit a negative trend (relatively more absorption).

Trends in scattering and absorption coefficients are also estimated by Mortier et al. (in review 2020) using AeroCom and CMIP6 models that have simulated the historical evolution of aerosol properties. For both variables, simulated trends can reproduce SARGAN derived trends suggesting significant decreases found over North America and Europe, although it must be considered that the number of models providing trends in σ_{ap} and σ_{sp} remains limited. Comparison with observations is also restricted to sites below 1000 m asl which further reduces data points for comparisons. However, decreasing trends in AOD and sulphate are observed for North America and Europe for both model and observational data. Asian in situ surface data are too sparse to derive a regional trend for that region but it is worth indicating that non statistically significant AOD and sulphate trends are found in the overall period 2000-2014 over southern and eastern Asia. This suggests that there are different trends in aerosol burden between North America and Europe and Asia. From model data alone, a global trend can be derived. Globally, the average model trend for 2000-2014 amounts to an increase of +0.2 %/yr for σ_{sp} and +1.5%/yr for σ_{ap}, respectively, higher than what is observed at ground-based stations.

There are some discrepancies between the work of Collaud Coen et al. (2020) and Mortier et al. (in review, 2020) in particular regarding trends derived for specific regions. This may result from different methods used to aggregate measurements to long time series, or to differences in the time period (2000-2018 versus 2009-2018) but, overall, they both confirm the shift of polluting activities from the developed countries to the developing countries during the last two decades and may also demonstrate the relatively higher reduction of BC-rich emission in some regions, which will affect aerosol forcing estimates.

The new figures are as follow

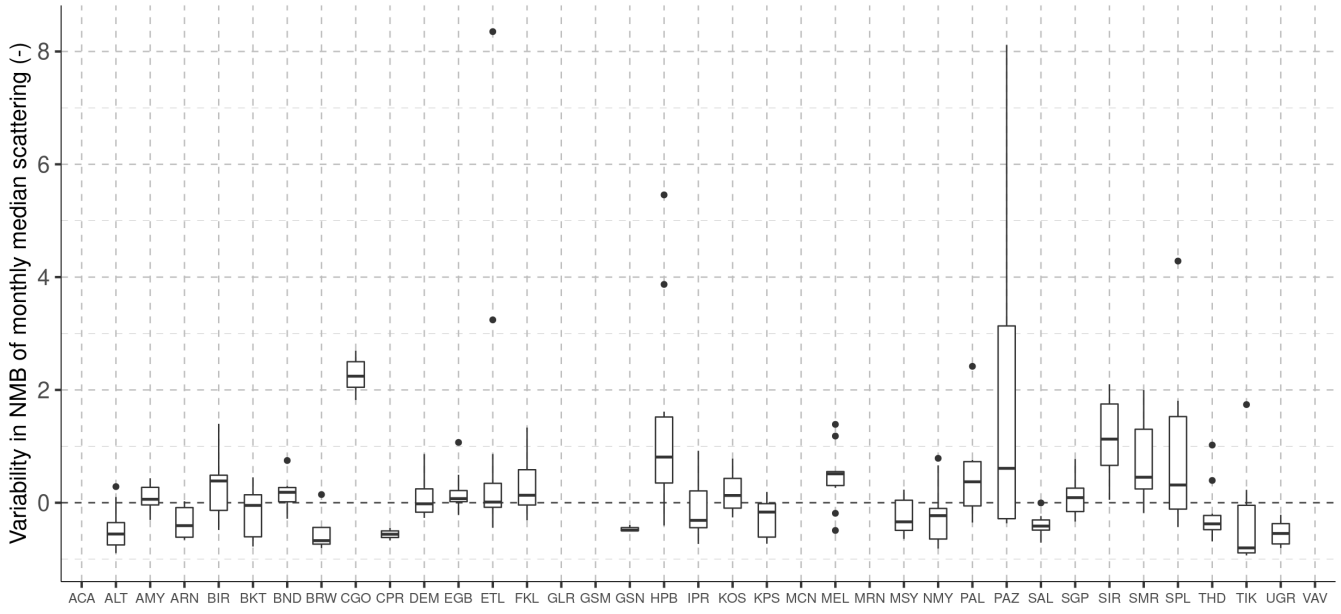

Figure 11a: Boxplots of the Normalised Mean Bias (NMB) for each SARGAN site, based on monthly median in-situ observation and the corresponding monthly median AeroCom simulation result. For scattering coefficient.

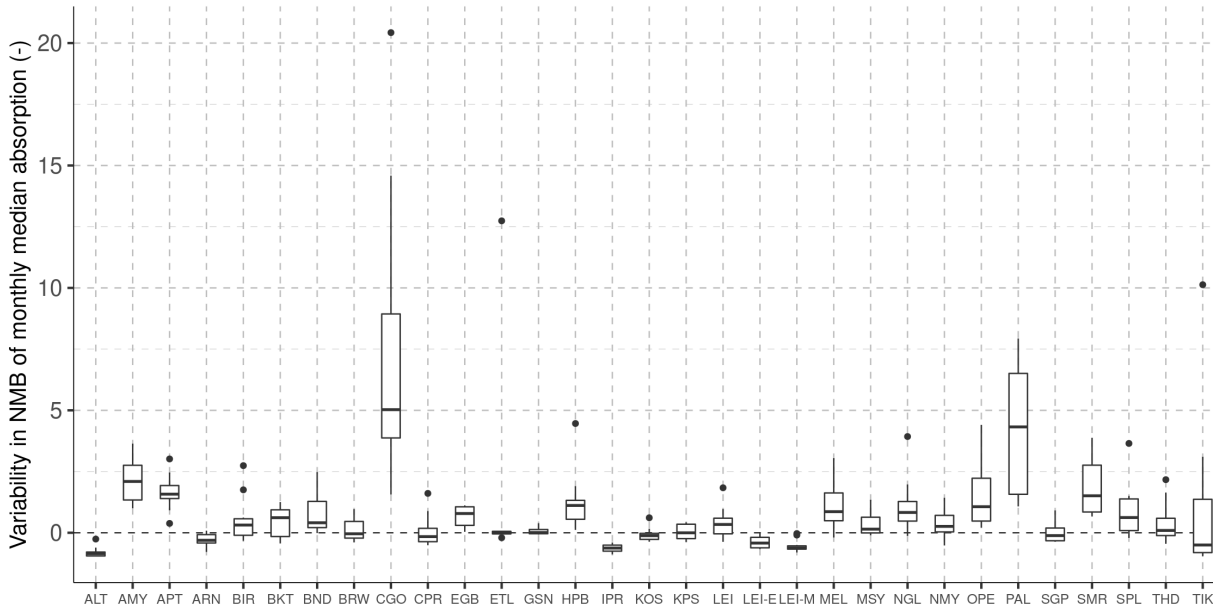

Figure 11b: Boxplots of the Normalised Mean Bias (NMB) for each SARGAN site, based on monthly median in-situ observation and the corresponding monthly median AeroCom simulation result.for absorption coefficient

Minor comments: L149. "45% of the variance": Do you mean here the inter-model variance? Please be more specific.
This refers to Carslaw et al., paper in 2013. It refers indeed to the variance of a multi-model ensemble when varying the reference year for the pre-anthropogenic emissions. This is now added to the new version.

L156. "important" here is too colloquial. Please remove or reword it.
« important » is now removed

Table 1. "Hyphen symbol" is misused as "Minus symbol" at several places in Table 1. Please correct.
We have checked and it seems that we are using the « Minus symbol » and not the « Hyphen symbol » throughout the Table. This could be rechecked during the editing process.

L270. Define the acronym "SARGAN" here.
This is now defined when first used

L358. Define "AE31" or refer to Table 2 here.
This is now corrected

L440. Is there any specific intention to use brackets around "product"?
The proper reference has been added to explain the use of brackets

L531. Define "N". I suppose it means particle number concentration.
N is now defined when first use

The section titles 6.2.1.~6.2.2 are missing. Please check.
This is now corrected