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

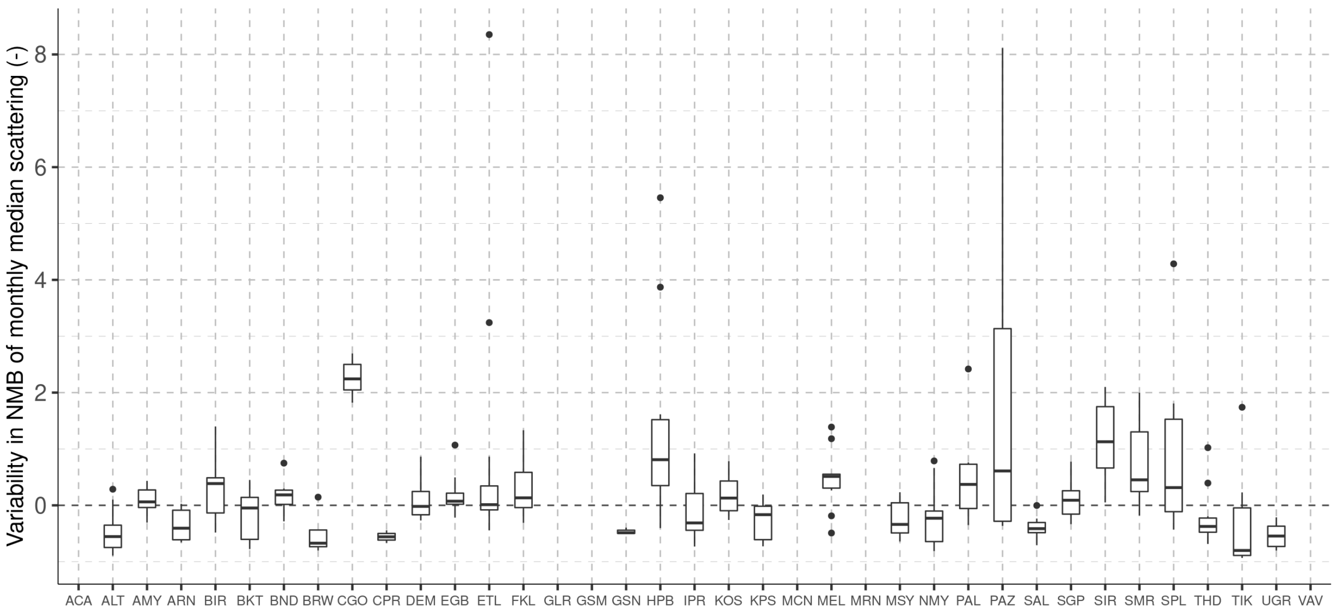

Figure 11a: Boxplots of the Normalised Mean Bias (NMB) for each SARGAN site, based on monthly median in-situ observation and the corresponding monthly median AeroCom simulation result. For scattering coefficient.

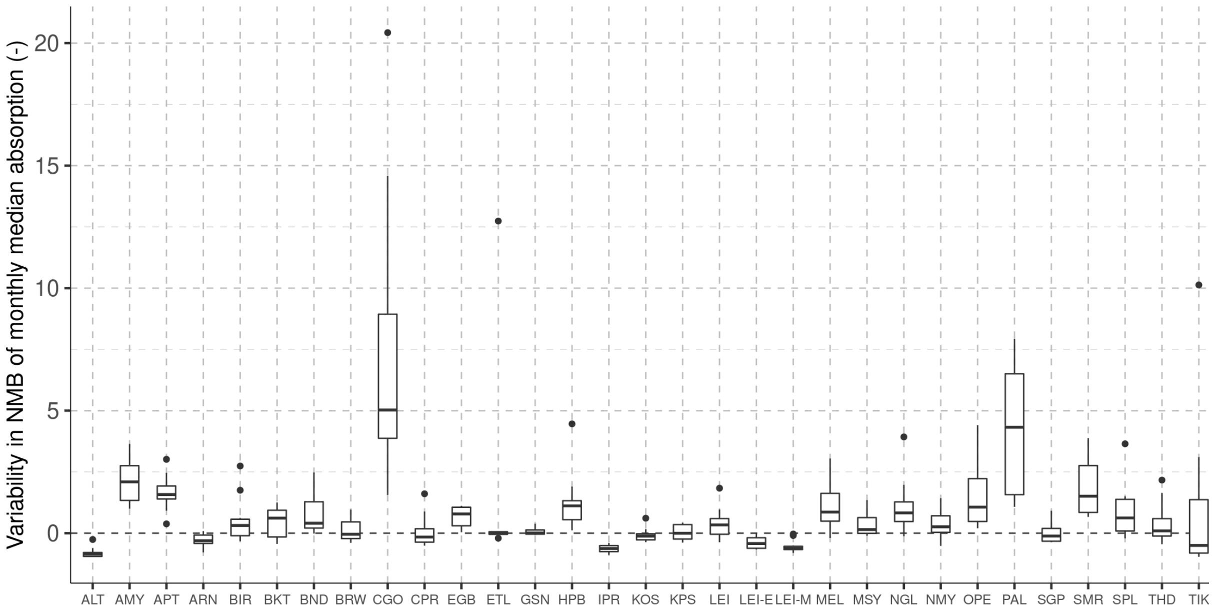

Figure 11b: Boxplots of the Normalised Mean Bias (NMB) for each SARGAN site, based on monthly median in-situ observation and the corresponding monthly median AeroCom simulation result.for absorption coefficient

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