ar-2025-12: "AIDA Arctic transport experiment (part 1): simulation of northward transport and aging effect on fundamental black carbon properties"

Answers of the authors to Reviewer#3

While the reviewer's comments are given in **black bold**, our answers are given below in blue letters. Additionally, we added the changes made in the revised manuscript in *blue italic letters*.

The manuscript titled "AIDA Arctic transport experiment (part 1): simulation of northward transport and aging effect on fundamental black carbon properties" presents a well-structured and insightful experimental investigation into black carbon (BC) aging under conditions representative of Arctic transport. To accurately quantify and understand the impact of atmospheric aging on BC properties and radiative forcing, the ARCTEx project simulated BC aging under quasi-realistic Arctic conditions in the AIDA. Informed by reanalysis data, four distinct scenarios were developed to capture seasonal and altitudinal variability during Arctic transport. The use of the AIDA chamber to emulate these variations is methodologically sound and lends credibility to the experimental design. The results on coating composition, morphological evolution, and aging timescales provide valuable empirical constraints for improving the representation of BC aging in atmospheric models.

We would like to thank the referees for their detailed and constructive comments, which helped us to improve our manuscript. Here we provide some major considerations. Several reviewers noted an insufficient discussion on the ambient representativity of transport conditions, soot generation and coating species. This was now addressed in more detail in both the Methods and Results sections, representing the major modification to the text. Reviewers also identified inconsistencies in the use of acronyms and abbreviations. These have been reviewed and corrected throughout the manuscript text and figures. The reviewer's specific comments are addressed as it follows.

Specific comments

While the dominance of nitrate and organic coatings is clearly demonstrated, the absence of sulphate or ammonium in coatings (Section 2.4.3) should be discussed. Clarifying this would help readers assess the generalizability of the findings.

This point was made by other reviewers as well, so the text was modified in several sections to better explain this point. We intentionally excluded sulfate, ammonia, and chlorine to focus on nitrate and organic ageing processes. Even with a simplified chemical system, ARCTEx showed that ageing under changing environmental conditions is still complex. This evolving-ageing setup raised additional questions, pointing to the need for further, focused experiments to better understand specific processes like organic and nitrate competitive oxidation, temperature-driven coating formation and inhibition of coagulation by coating. More detailed explanations are given as follows.

- The aim was to investigate coating with nitrate which have been often disregarded in the past. Hence, in the introduction we now describe the relevance of studying nitrate species and their interaction with BC: *… "BC variability in the Arctic was often associated with co-emitted sulfate aerosol from anthropogenic sources (e.g. Massling et al., 2015) and organic aerosol from biomass burning events (e.g. Moschos et al., 2022), while its correlation with nitrate was mostly ignored (AMAP, 2021). Similarly, chamber studies focused on the evolution of BC properties as function of internal mixing with sulfate (e.g. Möhler* *et al., 2005; Khalizov et al., 2009; Henning et al., 2012) and organics (e.g. Lefevre, 2019; Wittbom et al., 2014). As a consequence, the impact of BC-nitrate internal mixing on fundamental and climate relevant properties remained poorly assessed (Yuan et al., 2020). Internal mixing of BC with nitrate species becomes particularly important in the Arctic region, where, nitrate aerosol concentration have been increasing since the '80s despite an overall reduction of nitrogen oxides emissions (AMAP, 2021). The same report underlined how few studies focused on nitrate aerosol in the Arctic, introducing a knowledge gap on the sources of its precursors, its formation mechanisms and its interaction with other atmospheric species such as BC." …*

- The volatile organic aerosol, which may oxidize to secondary organic aerosol and condense on BC represented a non-controlled source of coating precursors, which is representative of the mini-CAST burner. This point was explicitly specified in Section 2.1.4 "Atmospheric composition":
  … "It must be noted that volatile organic compounds which are a by-product of combustion, were simultaneously emitted with BC and injected in the AIDA chamber without active control. As a result, the organic aerosol content in AIDA reflects the specific emissions of the burner and not ambient-like conditions. Therefore, although the experiments primarily targeted the evolution of BC mixing with nitrate coatings, the presence of organic vapours may interact or compete with NO_2 during condensation and coating formation, introducing additional complexity to the aging dynamics." …

- We did not observe sulphate or ammonia coatings because they were neither injected directly, nor co-emitted with soot by the mini-CAST burner. This is now stated properly in Section 2.4.3 "Chemical characterization of non-refractory aerosol particles":
  … "Since sulfate, ammonium, and chloride were not introduced into the AIDA chamber, directly or indirectly as a byproduct of combustion," …

Section 3.5: Briefly justify the use of the Hill equation for modelling aging timescales.

As also requested by Reviewer 2, more details on the Hill equations were added in Section 3.5 "Aging timescales":

*… "Next, we derived the aging timescale necessary to reach the critical coating mass for each scenario. We applied a Hill equation (Weiss, 1997; Goutelle et al., 2008) to model the non-linear relationship between Rm_{coat} and BC age, forcing the upper (maximum Rm_{coat} observed during WL) and lower (initial Rm_{coat} from each scenario) boundaries along with a 95% confidence interval (**Error! Reference source not found.**a, dashed lines). C ompared to a more generic sigmoid equation, the Hill function allowed capturing the sudden increase of Rm_{coat}, in the early phase of the ageing." …*

The manuscript could be further strengthened by discussing implications for global or regional climate models, particularly in light of recent developments in BC aging parameterizations (e.g., https://doi.org/10.5194/acp-25-2613-2025 and DOI:10.1029/2024JD041135).

- We really appreciated this comment, because it reinforces the novelty of our findings which fits in a recent set of publications raising an interest towards the importance of ageing timescale of BC. These works include Chen et al. (2024), Fierce et al. (2025) and Jin et al. (2025). Modifications were done in Section 3.5
- To reflect the finding of the Section, its title was changed from "Ageing timescale" to "Varying ageing timescales as function of transport pathway".
- A brief introduction to summarize how ageing is treated in models is now given at the beginning of the section:
  … "Global models parametrize this conversion in several ways. The most simplified approach in bulk aerosol models is to consider a fixed ageing timescale (Koch et al., 2009). Other aging schemes, used in modal aerosol modules, include a coating thickness threshold made up of a variable number of mono-layers (Liu et al., 2016). Considering the large discrepancies between observations and simulations, more detailed treatment of size, morphology and mixing is implemented in the modules of global models (e.g. (Matsui, 2016; Chen et al., 2024; Jin et al., 2025)." …

- We describe our findings in the context of modelling results at the end of the section:
  … "Our experimental results reinforce the recent findings of Fierce et al. (2025), who highlighted the inadequacy of fixed ageing timescales in models. Their work confirms that ageing rates are regionally and seasonally dependent, as observed in the ARCTEx scenarios, significantly affecting simulated BC concentrations, particularly in the Arctic. Moreover, the altitudinal and seasonal ageing patterns shown in ***Error! Reference source not found.*** *reflect ambient variability and lead to heterogeneous impacts on t he hygroscopic and optical properties of BC (Jin et al., 2025)." …*

Figure 3: Adding error bars or uncertainty shading would help visualize variability.

In the revised version of the manuscript, we have expanded the discussion regarding simulated vs. measured conditions. Overall, focussing on the NO2/BC ratio, we might have introduced some bias in the AIDA simulations compared to the ambient variability, especially in the low altitude scenarios. This is now discussed in Section 3.1 "Northward transport conditions". Following the request of multiple reviewers, we also modified Figure 3 adding the standard deviation of the reanalysis data and AIDA measurements. The modifications include: i) shading representing the standard deviation of both reanalysis and measurements; ii) different colouring to improve readability; iii) adjustment of axis labels and legend.

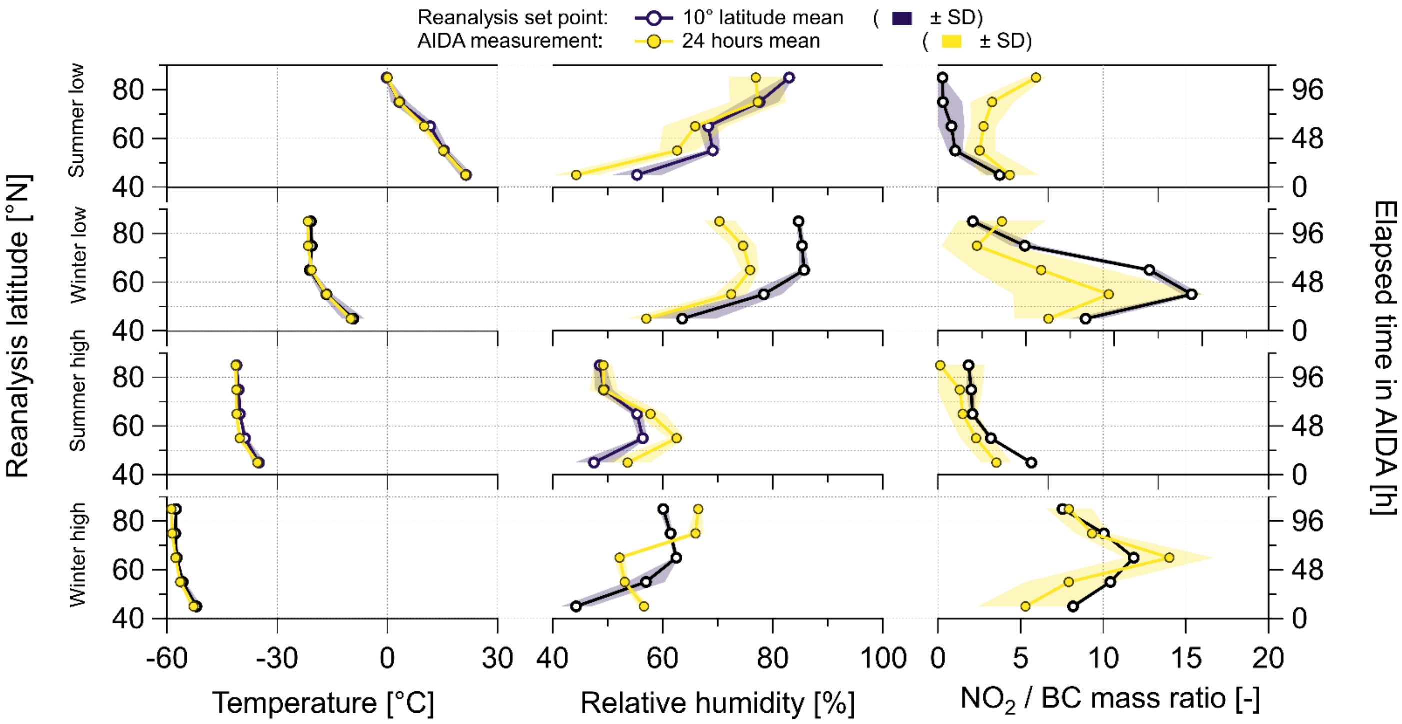

Figure 3 Left axis: Latitudinal profiles of temperature and relative humidity extracted from ERA-5 and of NO_2/BC mass ratio extracted from CAMS in the region of interest (40-90°N and 60-140°E). Mean and standard deviation (SD) calculated for equidistant latitude bands 10° wide. Right axis: temporal variability of temperature, relative humidity and NO_2/BC ratio measured in the AIDA chamber. Mean and standard deviation (SD) calculated over 24 hours.

Line 599 and 619: The unit "kg m^3" should be corrected to "kg m^{-3}".
Figure 8: The variable label "Rm$_{Coat}$" should be revised to "Rm$_{coat}$" to ensure consistency.
Table A1: Please verify that the list of abbreviations is complete. For example, "Rm$_{coat}$" appears in the manuscript but is missing from the table.
Ensure consistent use of notation, like kg m^{-3} in the manuscript and "Kg m^{-3}" in the Table A1.

We thank the reviewer for these comments. We verified the consistency of abbreviations and symbols in the manuscript and supplementary material and found many discrepancies. We now harmonized all abbreviations and symbols included in the text and figures according to the table in appendix.

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