Investigating the dependence of mineral dust depolarization on complex refractive index and size with a laboratory polarimeter at 180.0° lidar backscattering angle

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Submitted to Atmospheric Measurement Techniques (AMT).

We thank both reviewers for the time they spent to review our manuscript. The provided comments add value to our manuscript and may help future readers and we are grateful for that. For the sake of clarity, each comment is first recalled in italics, then our answer is given, together with a list of changes made to the manuscript, which also appear in red-lined version in the revised manuscript.

Author's response to Reviewer #2 's comments

Reviewer #2: *"This paper investigates the relationship between the particle depolarization ratio of mineral dust and the particles' complex refractive index and size, using a Pi-polarimeter that operated at a 180-degree backscattering angle, at two typical lidar wavelengths, 355 and 532 nm. Through laboratory experiments, the authors derive 16 dust-related particle depolarization ratio values that correspond to four different refractive indices (mineral dust samples with different mineralogy), for two size distributions (fine, coarse) and at two wavelengths (355, 532 nm).*

The work falls well within the scope of AMT. Overall, the methodology is well explained and the results are clearly presented. However, the manuscript could be improved prior to publication, by addressing the comments provided below."

We thank Reviewer #2 for the time she / he spent to review our manuscript. We are pleased to see that our work provides clearly presented results, in a well-explained methodology. For clarity, we answer point by point below to each comment. We thank Reviewer #2 for these improvements, which may help future readers. All the corrections made appear in red-lined version in the revised manuscript.

Major comments:

Reviewer #2, comment 1: "*The description of the dust samples (Sect. 2) should be more detailed. What exactly is Asian dust? Where do you get it from? Does it originate from a specific dessert? As I understand it, you use commercially available dust samples and silica and hematite as well. How are your four samples treated and prepared by the manufacturer? Furthermore, the paper would become richer, if the discussion about the mineralogical composition of desert dust samples could be added. There are various studies investigating the mineralogical composition including silica and hematite contributions. Apart from the finer/coarser SD differentiation (L144-L147), to which extent are the chosen dust samples representative/characteristic of what is being observed in the atmosphere?"*

- Answer to Reviewer's 2 comment 1: The Asian dust sample we use is provided by Powder Technology (commercial name: Kanto Loam). It was chosen for its large percentage in hematite (17-23 %), to investigate the role of the imaginary part of the refractive index on the PDR. It is commonly used as a test dust interferon in pollen light scattering measurements in Japan (Iwai et al., 2013), hence representative of observed atmospheric Asian dust. In this way, we symmetrized our approach by dealing with both Arizona Test Dust and Asian Test Dust. The information on the preparation of the samples is unfortunately not provided by the manufacturer. Nevertheless, the measured SD are representative of what is observed in atmosphere, with a low number concentration of more than 10 µm particles, as observed by Weizierl et al. (2017), and as detailed in our answer to Reviewer #1. Also, our SD correspond to that currently observed in other laboratory experiments (see Järvinen et al., 2016 for example).

The mineralogical composition is only given for hematite and silica, which are the main concern of our study. Other chemical oxides are also present in our dust samples in various percentages but their mineralogical composition is not detailed since they exhibit negligible imaginary parts of CRI compared with that of hematite. Also, as explained at lines 398-400 and 439 of our original manuscript, studying their PDR is an outlook of this work. To prepare this future work, we can of course add this information in % of Arizona Test Dust, then Asian Dust: Al_2O_3 (11 %, 29 %), CaO (4 %, 1.5 %), K_2O (3.5 %, 0 %), Na_2O (2 %, 0 %), MgO (1.5 %, 5 %), TiO_2 (0.5 %, 2 %). Thank you for your comment.

- **Changes made to the manuscript:** To account for Reviewer #2's comment, we added the following sentences to our revised manuscript:
  - In Section 2.1, dedicated to the presentation of our dust samples: "For Asian dust, we use a commercial sample provided by Powder Technology (commercial name: Kanto Loam), commonly used as a dust interferon in pollen light scattering measurements in Japan (Iwai, 2013), hence representative of observed atmospheric Asian dust. In this way, we symmetrized our approach by dealing with both Arizona Test Dust and Asian Test Dust."
  - The information on the mineralogical composition of our test dust samples was also added: "Other chemical oxides are also present in our dust samples in various percentages, but with negligible imaginary parts of CRI compared with that of hematite. Investigating the PDR of these oxides is then beyond the scope of this paper. Their percentage in (Arizona Test Dust, Asian Dust) is given for clarity: Al_2O_3 (11 %, 29 %), CaO (4 %, 1.5 %), K_2O (3.5 %, 0 %), Na_2O (2 %, 0 %), MgO (1.5 %, 5 %), TiO_2 (0.5 %, 2 %)."
  - In Section 2.2, dedicated to the size of our dust samples: "The measured SD are representative of what is observed in atmosphere, with a low number concentration of more than 10 µm particles, as observed by Weizierl et al. (2017)".

Reviewer #2, comment 2: "*The size distributions (Fig. 1) are a finer and a coarser one as you often state, but it is not a fine mode and a coarse mode (as sometimes ambiguously stated, e.g., L12, L426). It is a fine mode size distribution and a fine + coarse mode size distribution or in other words a size distribution with and one without coarse mode. Please clearly make this statement in Sect. 2.2. »*

- **Answer to Reviewer's 2 comment 2:** Thank you for your comment, which adds precision to our manuscript. We agree and modified our manuscript accordingly.
- **Changes made to the manuscript:** We added this sentence to Section 2.2: "More precisely, the two considered SD correspond to a size distribution with and without coarse mode".

Reviewer #2, comment 3*: "How do you estimate the uncertainty of your results (Tab. 1 + 2)? Is it the uncertainty of the fit? To which amount does the systematic error is considered?"*

- **Answer to Reviewer's 2 comment 3:** Thank you for your comment. We indeed precisely evaluate the uncertainties in Tables 1 and 2 by applying the least-square fit method to the measured light backscattering light intensity. The systematic errors are minimized as explained in the beginning of Section 3.5, so that corresponding uncertainties are mainly due to statistical errors.
- **Changes made to the manuscript:** To account for Reviewer #2's comment, we added the two following sentences to the caption of Table 1: "The uncertainty on $F_{22,\lambda}/F_{11,\lambda}$ is deduced from the evaluation of b_λ/a_λ, itself deduced from the least-square fit adjustment of I_λ. The uncertainty on $F_{22,\lambda}/F_{11,\lambda}$ is mostly dominated by statistical uncertainties since our biases are minimized, as explained in Section 3.5."

Reviewer #2, comment 4*: "The discussion and comparison to previous literature is rather short and should be extended before publication. Even if previous laboratory setups did not operate at exactly 180°*

backscatter, the results should be discussed. Especially, I am missing a reference and discussion to the work by Sakai et al., 2010, who investigated fine and coarse mode dust from Asia and the Sahara at 532 nm. How do their results compare to your new findings? The comparison to lidar field experiments is rather short as well. It is hard to compare for Arizona Test Dust, but for Asian dust, there are plenty of field experiments reporting PDR at 355 and/or 532 nm, e.g., Sugimoto & Lee, 2006; Hofer et al., 2020 or Hu et al., 2020."

- Answer to Reviewer's 2 comment 4:

  Thank you for your comment. As you noted, comparison with other laboratory work at strict backscattering angle of 180.0° is not feasible since the scattering matrix elements may strongly vary over a small scattering angle range, especially when light absorbents are present (Cholleton et al., 2022). Otherwise, according to Eq. (3), a small variation in $F_{11,\lambda}$ and in $F_{22,\lambda}$ can result in a large variation of the lidar PDR. Though our samples are different, Sakai et al., (2010) retrieved increasing lidar PDR with size at 532 nm wavelength, what we also observe. We already compared our results with Hofer et al. (2020) at line 370 of our original manuscript and found an increase in the dust lidar PDR with particles size. We may precise the values: Hofer et al. (2020) reported lidar PDR ranging from 0.23 to 0.29 at 355 nm wavelength, in line of our laboratory results. The differences are more pronounced at 532 nm wavelength. Comparison with lidar field experiments, involving particle mixtures, with a more complex distribution of sizes and refractive indices, is however not straightforward, as underscored by comparison with Hu et al. (2020) who reported 0.28-0.32 ± 0.07) at 355 nm wavelength.

  Changes made to the manuscript: To account for Reviewer #2's comment, we added the above discussion to our revised manuscript and especially reference to Sakaï et al. (2010) at line 370 of the original manuscript where we emphasized that the PDR was increasing with particles sizes. We also added that "Comparison with lidar field experiments, involving particle mixtures, with a more complex distribution of sizes and refractive indices, is however not straightforward, as underscored by comparison with Hu et al. (2020) who reported 0.28-0.32 ± 0.07 at 355 nm wavelength."

Minor comments:

"Please always state Arizona Test Dust and not just Arizona dust. Arizona Test Dust is a well-known term in the community."
Thank you for your comment, we modified the manuscript accordingly.

"L43-47: need rephrasing. Also, the literature selected is rather limited, important studies are missing."
We will add reference to Hofer et al. (2020) and Hu et al. (2020) there in the revised manuscript.

"*L49: The particle linear depolarization ratio's importance for aerosol typing has been demonstrated in numerous studies (e.g., Burton et al., 2012). The authors should extend the literature provided here accordingly."*
We already quoted the paper by Burton et al. (2016), but we can of course also add that from 2012.

"*L70- 82: It would have been better if the authors merged the list with the main body text."*
We do not want to merge the list with the main body text as it is important to emphasize the novelty of our work compared with that of other existing laboratory experimental set-ups.

"*L102: new paragraph "The paper is structured…"*
We agree and did the correction.

"*L122-124: The imaginary part of the CRI varies by a factor of 10 between the literature values: 0.0925 (Longtin et al., 1988) and 0.9 or 0.6 (Go et al., 2022). Is there a reason for the difference?"*

Thank you for your comment. Triaud et al. (2005) is in agreement with Go et al. (2022). We will keep Go et al. (2022).

"*Just out of curiosity: Why do your size distributions (Fig. 1) all show a peak at 1 µm?"*
This is a good question: as we did not yet explore all size distributions, it is difficult to explain. It however remains representative of observed atmospheric mineral dust size distributions.

"*In line 267 you're referring to the polarization lidar reference paper of Freudenthaler et al., 2009. There is an even more complete assessment of the polarization lidar calibration given by the same author (Freudenthaler, 2016). There, additional sources of uncertainties are discussed. In your case, the rotational misalignment around the optical axis might be worth discussing (even if it is probably very small)."*
We will add reference to Freudenthaler et al. (2016) at line 267 of the revised manuscript. We also checked that the rotational misalignment around the optical axis is very small.

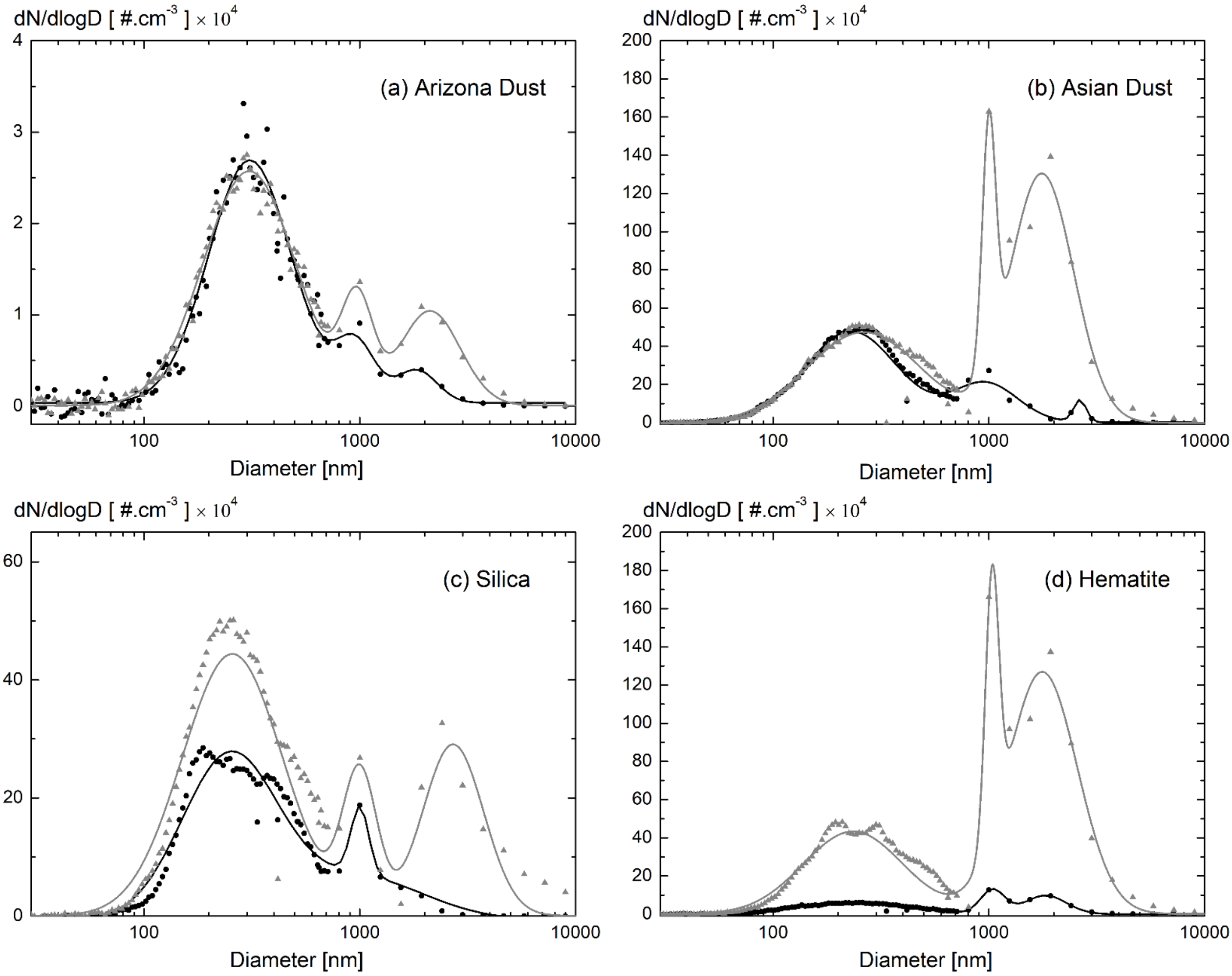
"*L282-284: Please provide an approximate particle concentration."*
The concentration can be found be integration over the Figure 1 size distributions. We found 1.11×10^3 cm^{-3} for the coarser SD and added the value to the manuscript.

"*Lidar particles depolarization ratio – lidar PDR: Does the term "lidar PDR" refers to the 180° backscatter direction? Or what is the difference to PDR?"*
Thank you for your comment. In Equations (3) and (4), we precisely emphasized the distinction between PDR and the lidar PDR, the latter being specifically dedicated to the 180.0° lidar backscattering angle.

"*At one instance, you should mention that you are measuring the linear depolarization ratio."*
We agree and added the word linear at line 186 and the following sentence to Section 3.1: "The PDR stated in Equation (3) is the linear PDR, which can be related to the circular PDR if need be (Mishchenko et al., 2002)."

"*L351-353: Please rephrase. In field experiments, we do observe pure aerosol conditions with lidars- not only aerosol mixtures."*
We agree and modified the sentence as follows : "Although in such lidar field experiments, the measured PDR is usually that of dust mixtures (Miffre et al., 2011), the comparison with our laboratory findings remains interesting."

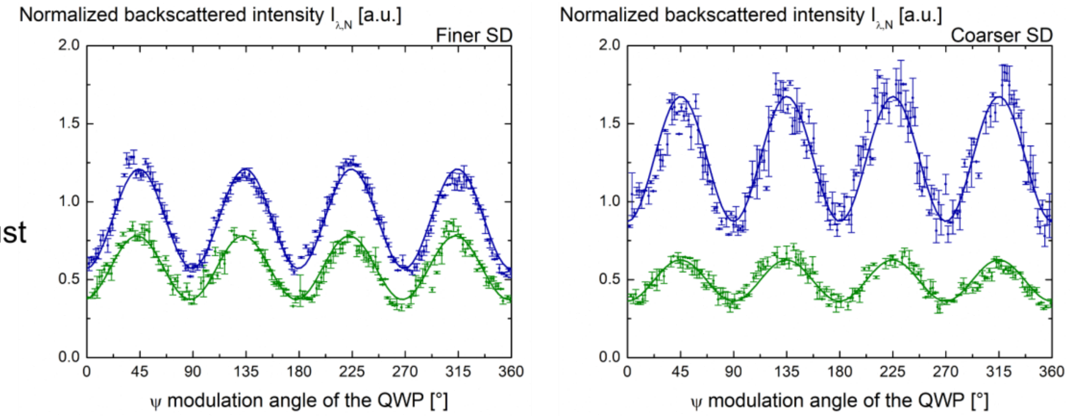
"*Fig. 1: Larger fonts (for labels, markers, axis) are needed. Consider changing the grey colour, it is very hard to read."*
Thank you for your comment, we did the proposed correction.



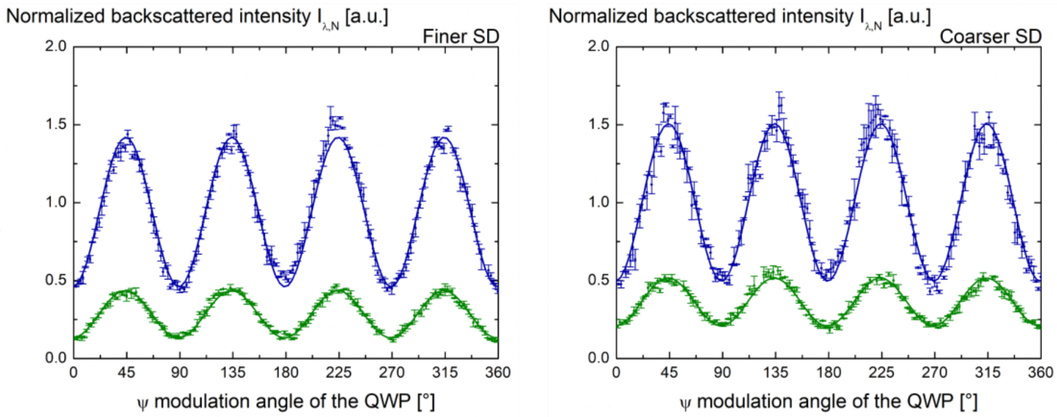
"Fig. 4: Larger fonts are needed. Very hard to read. There is enough space in the plot to include the names of the dust samples (Arizona Test, Asian). The same holds for Fig. 5."

Thank you for your comment, we did the proposed correction.

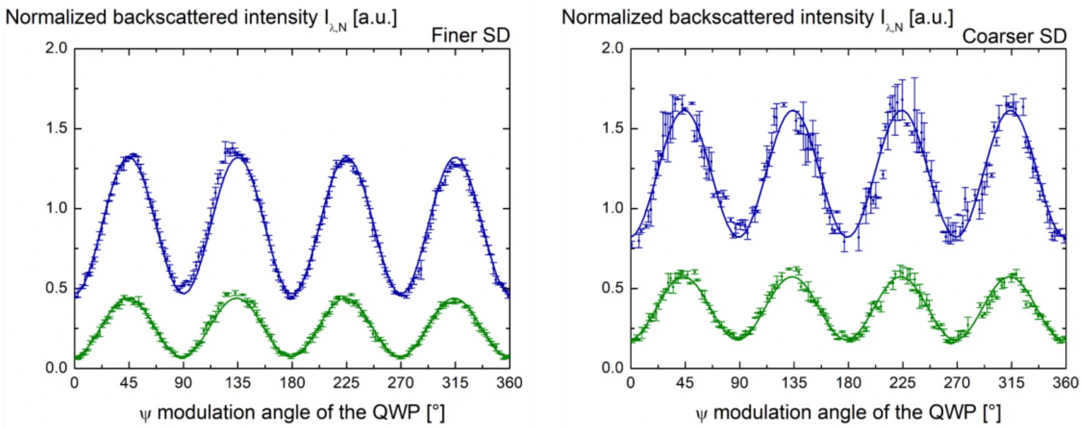
(a)
Arizona Dust



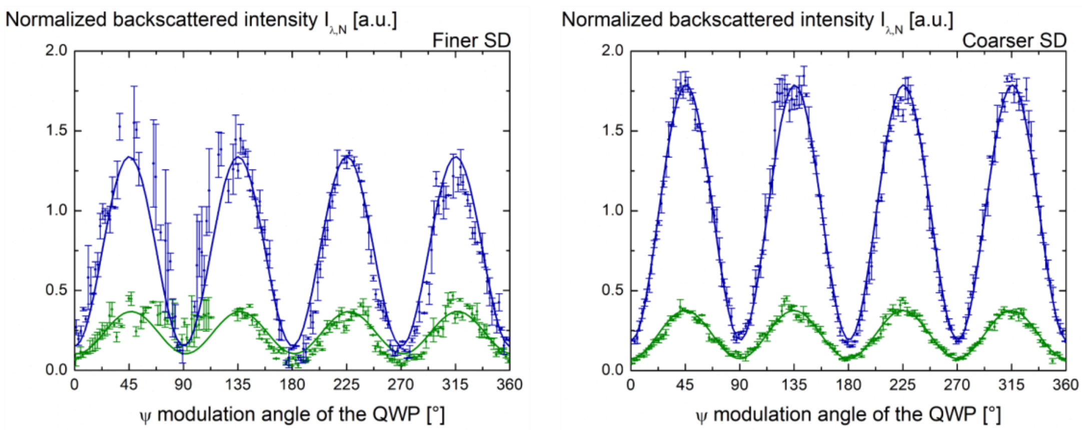
(b)
Asian Dust



(a)
Silica

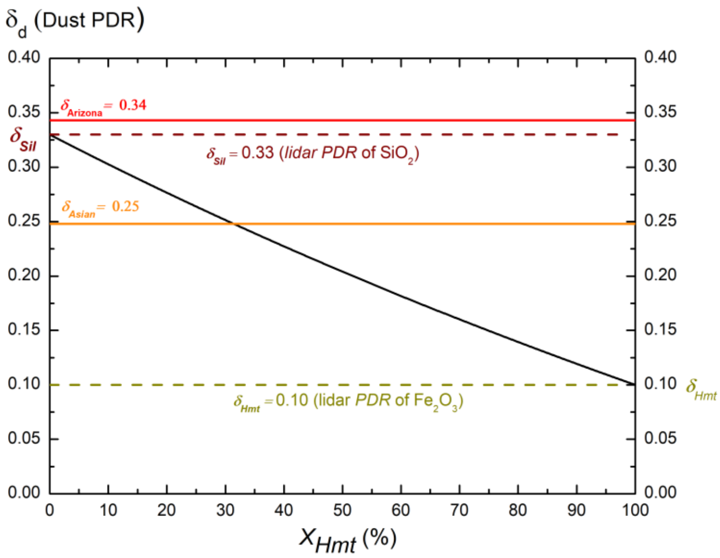


(b)
Hematite



"Fig. 6: It would be recommended to insert the results for Asian dust and Arizona Test Dust into the figure. Even if they are not lying perfectly on the line, it illustrates better the consistency of your results. By the way, the information about the depolarization ratio of silicate and hematite is doubled (once next to the figure and once on the dashed line)."

Thank you for your comment, we plotted the proposed correction as shown below. While the dust PDR should lie in between δ_{Sil} and δ_{Hmt}, we observe that δ_{Sil} then becomes lower that $\delta_{\mathrm{Arizona}}$. Since both Arizona and Asian dust are complex mixtures of more than two components (Sil, Hmt), their PDR may exceed that of pure components (δ_{Sil} and δ_{Hmt}) as is the case here for Arizona. It is not surprising as the fraction of hematite is low in ATD, and so the potential effect of other components is more visible. As a result, the figure we present below may confuse future readers. To improve the readability of our contribution and avoid confusion, we then chose not to add $\delta_{\mathrm{Arizona}}$ and δ_{Asian}. To compare a mixing rule with laboratory evaluations, we should consider more components mixtures, which is beyond the scope of this study, as stated in our manuscript. Thank your for the proposed improvement on the repetitive information on the PDR, which we corrected.



"Eq. 8: Indices should not be in italic."
Thank you for your comment, we did the correction.

"The figures should be provided in higher resolution, with larger fonts. In their current state, they are very difficult to read."
Thank you for your comment, we did the correction.

"Sections 4.3 and 5 are rather repetitive. I suggest merging those sections into one to avoid text repetitions."
Thank you for your comment. In line of our answer to Reviewer #1's comment on the same topic, we rather chose to rename the title of Section 5 to "Summary and conclusion". We expect it will help future readers.