In this manuscript, the authors investigate the light absorption characteristics of solvent-extractable brown carbon aerosol in ambient samples affected by multiple sources. The manuscript raises the topic of the effect of the selected solvent, or solvent mixture, on the extraction efficiency of organics and subsequently on the absorbance measured by UV-visible spectroscopy and associated light absorption properties (mass-normalized absorbance and its wavelength dependence). They find that N, N-dimethylformamide dissolves BrC associated with unburned fossil fuel and polymerization processes of aerosol organics more efficiently than methanol. The study results and potential implications are of interest to readers of *Atmospheric Chemistry and Physics.* Nevertheless, I would only recommend publication of this manuscript upon careful consideration by the authors of the specific comments below and subsequent clarifications and fine-tuning of some discussions in the revised manuscript.

Specific comments

Line 1: the message of the title may not be universal and could be considered misleading. If the authors prefer the current title to largely remain (although Reviewer 1 has already provided an alternative that may better reflect the paper content), I strongly recommend they change it to: “Potential underestimation…”. The main reason is that methanol still efficiently extracted biomass burning-related BrC, which has been more widely studied in the literature.

Lines 44-45: for completeness, please clarify the following details in the abstract: (1) BrC aerosols associated with biomass burning, and coal (?), combustion sources were still highly soluble in MeOH; (2) the different MeOH solubility of BrC from different (seasonal) sources was likely the main reason for the aforementioned distinct time series. Therefore, (3) a more accurate alternative to the sentence: “These results highlight the necessity of replacing MeOH with DMF for further investigations on structures…” could be: “These results highlight the importance of testing different solvents to investigate the structures and light absorption of BrC, particularly of the low-volatility fraction potentially associated with certain non-traditional sources.”. Please also rephrase related statements in Lines 103-104, 105-107, 227-228 (“potential underestimation”), 415, 418 (“may sometimes”), 423-424 (“…may potentially underestimate the contribution of solvent-extractable…”), and elsewhere if applicable.

Line 93: the sentence is more informative with the following (or similar) addition referring to the extractable aerosol fraction: “…directly if the latter is not converted to particulate absorption with Mie calculations, solvent-matrix, and pH effects are not accounted for, and solvent solubility is not high.”.

Line 120: that is likely true for DMF; THF has been tested for biomass burning-influenced ambient BrC (Moschos et al., 2021); do the two observations agree? The authors state “rarely” in this sentence: do any other studies exist that have tested any of these two solvents for extracting BrC aerosol?

Line 195: when measuring the absorbance of solvent extracts, solvent-matrix effects (Reichardt, 2003) are not uncommon (yet rarely accounted for in the BrC research). Chen and Bond (2010; cited in the preprint), Mo et al. (2017), and Moschos et al. (2021) observed higher absorbance of water-extracted BrC aerosol that was further diluted/re-dissolved in methanol (for the same total extract volume). Could the authors discuss, in the revised manuscript, similar effects for their selected solvents/mixtures, as well as the implications for the results presented here when not correcting for such (i.e., currently, the solvent-matrix vs. solubility effects are not decoupled)? Can the authors rule out a solvent-matrix effect that would affect the wavelength-dependent comparison between MeOH and DMF, for example, in Fig. S4?

Line 312: here, the authors have the opportunity to discuss also potential pH effects, e.g., the absorbance red-shift for the water-extract of 4-nitro-catechol in Fig. S5 and the isosbestic point ~365 nm, which seem to be consistent with the observation of Lin et al. (2017) for water vs. organic-solvent BrC aerosol extracts.

Line 426: please clarify: "…in cold periods, when coal/biomass burning sources dominated the aerosol emissions..." if that is the case.

Conclusions and implications section: It is important here to provide a broader view that will allow future studies to confirm these observations, while other approaches may still be helpful: for example, the authors could state that a combination of solvents with a broad polarity index (e.g., Lin et al., 2018) may still be good choice to cover different conditions, e.g., a mixture of non-polar (e.g., hexane), polar protic (e.g., MeOH) and polar aprotic solvents (e.g., DMF) for a range of BrC-containing samples influenced by different sources. Based on this and other comments above, there is no "universal evidence" from this study that DMF is the unique-best solvent for BrC under all conditions. At the same time, a more balanced discussion in the revised manuscript would encourage future studies to test DMF and potentially verify or revise the authors' observations.

Line 434: does that refer to the isoprene oxidation factor in Fig. 3? That is an important finding; the figure can be cited once more in this section together with this statement.

Figure 3: please mention (possibly in the caption) that the biogenic emission factor *Abs* is below the detection limit (if that is the case). Further, I agree with Reviewer 2 that it is critical to provide evidence for the robustness of the PMF solution for a reader to assess the quality of the results and the validity of the associated conclusions. Could the authors also discuss the yearly evolution of the *Abs* relative difference between the two solvents for each PMF factor in Fig. 3? The relative difference seems low for coal and biomass burning throughout the year; what are the time-series trend and day-to-day variability for the other factors (those where both solvents seem to dissolve a non-negligible fraction of their chemical constituents)? Finally, based on the statement in Lines 297-299, could the authors reproduce Fig. 3 for *Abs* at a longer wavelength and compare the two PMF results?

Technical corrections

Lines 281, 295, 315 & 416: "low-volatility".
Table 3: correct the superscript to "Mm^{-1}".
Table S3: please provide the units of the tabulated data other than Abs_{365}.

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Lin, P., Bluvshtein, N., Rudich, Y., Nizkorodov, S. A., Laskin, J., and Laskin, A.: Molecular chemistry of atmospheric brown carbon inferred from a nationwide biomass burning event, Environ. Sci. Technol., 51, 11561–11570, https://doi.org/10.1021/acs.est.7b02276, 2017.

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