Author's comments

Key:
Referee Comments
Authors Response
Evidence in text of paper changes

The authors would like to thanks all 3 referees for insightful comments that helped improving the paper. Particularly, Pr Hans Puxbaum provided a long, precise, and detail text with account of the historical aspects of past research on cellulose measurements in atmospheric PM, which is extremely interesting and complete. His comments are now partially reflected in our paper, but the original texts (RC1 and RC4) should be referred to, in order to give him full credit on this point.

Response to Referee #1 (Hans Puxbaum): RC1 and RC4

General comments

The submitted paper constitutes a fine research work about a seldom reported topic: "Cellulose in atmospheric particulate matter".

The general appearance is excellent, there are clear intentions, interesting conclusions, and an intention to deliver a "new standard" in this field is glaring. In fact, the delivered data is one of three of the larger data sets obtained for the occurrence of atmospheric cellulose so far. However, the treatment of the past literature and the presentation of data leaves to be improved.

Historically the first large data set originated from the CARBOSOL Project – with data from 6 – regional to remote background sites in a west-east transect from the Azores to Hungary, spanning a two-year period, and published in a JGR series of papers overviewed by Legrand and Puxbaum (JGR 212; 2007; https://doi.org/10.1029/2006JD008271), and for cellulose reported by Sanchez-Ochoa et al. (JGR 212; 2007) – cited in the submission.

Second is the AQUELLA data set, from urban and regional sites in Austria. It comprises a large data set from several local and regional projects in Austria. The results were given in Reports to the Country Governments, however not published in scientific literature, except for following 10 sites: - 4 sites in Vienna, 3 sites in Graz and surrounding, and 3 sites in Salzburg and surrounding, with data over a year – these data are reported in detail in Alexandre Caseiro´s doctoral thesis (2008) – cited in the submission.

The third larger data set arrives now in the current submission, with data from 9 sites in France and Switzerland, over time spans of around two years.

There are also reported atmospheric cellulose data from shorter studies, including the primary paper on cellulose in the atmospheric aerosol (Kunit&Puxbaum 1996), and about the occurrence in fine particles (Puxbaum&Tenze-Kunit 2003). Followed by papers from measurement campaigns with aerosol-mass-spectroscopic instrumentation designed to find

more conveniently instrumentally accessible markers for plant debris or vegetative detritus (two papers from Yttri et al., cited in the submission, and papers from campaigns of the Paul Scherrer Institute – not cited in the submitted text (e.g. Lanz et al., EST 2008 - https://doi.org/10.1021/es0707207 ; Daellenbach et al., ACP 2017 - https://doi.org/10.5194/acp-17-13265-2017).

A clever presentation and comparison of recent data with earlier ones is presented in the (cited) Bozetti et al., EST 2016 paper, which seems to be a sort of precursor of the current submission. It is recommended to the current authors to think of a presentation for comparing cellulose data from different studies as shown in the Bozetti paper / or in another overview type graph or table.

Overlooked has been a quite important paper from the Aveiro Group about the Indoor-occurrence of particulate cellulose: Cerqueira et al. Atmos. Env. 2010 - doi:10.1016/j.atmosenv.2009.11.043.

And – there is a report about cellulose in Beijing aerosol determined with the Kunit-Puxbaum assay: Yi DING, et al., Study on the Vegetative Detritus Contribution to Beijing Urban $PM_{2.5}$ Using Cellulose as a Marker[J]. *Rock and Mineral Analysis*, 2013, 32(5): 738-746 (no doi given, access via Google scholar).

I absolutely recommend publishing the paper, however after a thorough revision including following improvements and corrections.

In the introduction various aspects of aerosol characterization are mentioned, of which most are only weakly related to the topic of determining an insoluble component of the organic aerosol. The background to understand the composition of insoluble organic particles and the contribution of vegetative detritus to the atmospheric aerosol dates back to the times of Glen Cass, Monica Mazurek, Lynn Hildeman, Berdt Simoneit, and Wolfgang Rogge, who was the thesis candidate and paper lead author. The primary point of the Rogge et al., Atmos. Env. 1993 - https://doi.org/10.1016/0960-1686(93)90257-Y paper for the cellulose issue was the clear statement that in the investigated case of LA aerosol, more than 50% of the organic material was insoluble in water and common solvents. And they proposed markers obtained from the “soluble” part to draw conclusions about insolubles, e.g. plant waxes for vegetative detritus – Rogge et al. EST 1993 - https://doi.org/10.1021/es00049a008. The vegetative detritus marker in the Rogge et al paper, however was going back to Simoneit and Mazurek`s paper in Atmos. Env. 1982 - https://doi.org/10.1016/0004-6981(82)90284-0. The search for primary biological particles was getting directed towards insoluble bio-polymeric material already by Matthias-Maser&Jaenicke, continued for cellulose containing particles by Kunit and Puxbaum.

The author’s would first like to address our very warm and deep thanks to Prof Hans Puxbaum for his high appraisal of our work, his very insightful comments, particularly on putting the studies of atmospheric cellulose in an historical perspective, and also (of course) for the prior contributions of his group in this field of research, which indeed form the basis of our work.

His synthesis (here and below, in RC1 and RC4) of the previous literature on the subject cannot be done better, and these texts are the most valuable introduction to our paper. We included part of the refences in the introduction and in the text of our paper (particularly lines 97 –

126 of the revised ms), but did not wanted to plagiarize his texts which are a model of an historical perspective.

Proposed improvements and correction (1 and 2)

"The treatment of the insoluble particles in the manuscript should disentangle the bacteria/fungal spores story versus the plant fragments story – while for the determination of primary "idividua" a range of possibilities is available, as is most of the literature about PBOP, for the plant fragments only a few groups are currently involved (see above)."

We have attempted to separate out the aspects relating to plant debris from fungal spores etc. and have included the fundamental literature suggested by Prof Hans Puxbaum.
Correction 3:

"Please sort the citations in text chronologically - by the year of appearance, then the primary information should get credit."

"In the majority of studies, at most 20% of the OM can be speciated and quantified at the molecular level (Michoud et al., 2021; Alfarra et al., 2007)" Actually, the sentence is sort of textbook knowledge, now what is the reason, to cite these two papers here? Did they increase the per cent output of OM?

Why we should think of the unability for GC-MS or aerosol-AMS techniques to find more of the soluble OC part, when the study is directed to the insolubles?"

All citations have been placed in chronological order. Michoud et al. (2021) found that 24% WSOM could be speciated when coupling the TD-GC-MS to a PILS-TOC (Particle Into Liquid Sampler – Total Organic Carbon). Without this, only 18% OM on average can be speciated. We feel like this is an interesting development, however the main thrust of this point was to highlight the complexity of characterising OM composition for context.

Specific points

Line 24: "the expression "a new method" should be reconsidered."

"Novel" method has been revised

"The spatiotemporal variations of free cellulose concentrations in atmospheric particles, as a proxy for plant debris, were investigated using an improved protocol with HPLC-PAD measurement."

Line 47-49: "What actually had been "deeply improved"?"

The author's feel that this work has investigated in the most depth this fraction of plant debris. For example, interannual variability of plant debris had not been investigated to the same degree – which has inadvertently led to an investigation of the impact of the COVID-19 pandemic on plant debris emission sources (more investigation is necessary to deconvolute this topic). Further, whilst relatively simplistic, investigations of cellulose with other chemical tracers (over a long period of time when meteorology is changing) using a large array of samples provides a relatively robust indication of co-emission. This is also the case for indicating the highly local nature of cellulose sources (via correlations of cellulose concentrations between sites over a long period of time). We hope that refinements and

revisions of this technique will arise over the coming years to aid both field and computational/PMF studies.

Line 55-57: "Primary citation on the EC/OC content of aerosols would be Novakov/Hansen (Designers of the Aethalometer) and Birch&Carey (Designers of the Sunset Instrument)."

Citation for Hansen/Novakov has been added for the EC/OC content of aerosols.

"Particulate matter is made up of elemental and inorganic material, as well as a significant proportion of material of a carbonaceous nature (organic carbon, OC, and elemental carbon, EC) (Hansen et al., 1984; Birch and Cary, 1996; Putaud et al., 2004a; Yttri et al., 2007a; Franke et al., 2017)"

"Organic carbon (OC) and elemental carbon (EC) were analysed with a Sunset Lab analyser following the EUSAAR2 thermo-optical protocol (Hansen et al., 1984; Birch and Cary, 1996; Aymoz et al., 2007; Cavalli et al., 2010)"

Line 75-76: ""solid airborne particles derived from biological organisms, including microorganisms and fragments of biological materials such as plant debris and animal dander" (Fuzzi et al., 2006; Després et al., 2012). You use a direct quotation and 2 citations – so who of the two stated the sentence?"

Fuzzi et al. (2006) has been removed from the script.

Line 83-85: "Which of the published emission estimates differentiated between viable particles and plant matter?"

Taken from Jaenicke (2005): "Our estimate of the strength of the 'source biosphere' for atmospheric primary particles, based on observed concentrations, the strength of other sources, and atmospheric residence times, is presently roughly 1000 Tg/year. Earlier estimates reflected a limited view of aerosolized biological components and were focused on organic aerosols".

Line 101: ""Cellulose is used as a molecular marker in order to quantify the total ambient concentrations of plant debris (Sánchez-Ochoa et al., 2007; Butler and Bailey, 1973)": Butler and Baily are a plant physiology textbook and for sure never stated, that cellulose might be used as marker in atmospheric studies."

Butler and Bailey (1973) has been removed.

Line 104: "What is meant by an "insufficient ambient condition"?"

This line was intended to show the scarcity of cellulose measurement studies, which tend to cover either rural or urban locations but rarely both (Caseiro (2008) aside). Rural studies have been primarily completed, and so our knowledge of cellulose in urban environments is even more limited.

This line has been changed to: "The number of campaigns investigating measurements of atmospheric cellulose are scarce in comparison and do not sufficiently cover all ambient environments".

Line 114: "There is a misprint: the delignification step for plant particles was first described by Kunit and Puxbaum 1996, following and down-scaling a method of Gould et al., 1984, for delignifying agricultural residues."

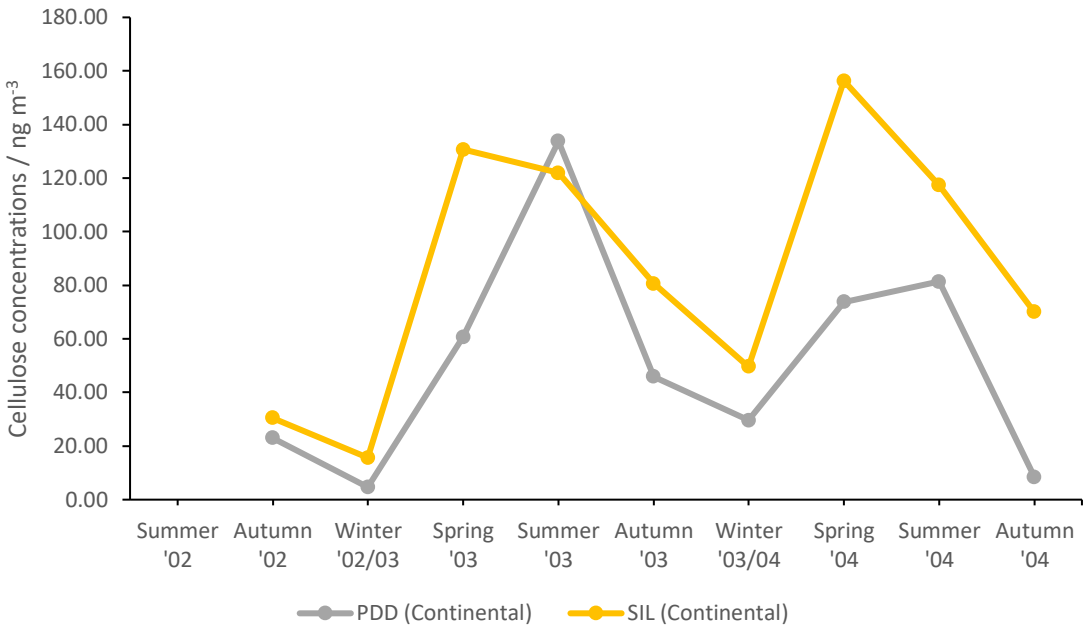
References have been added to provide the necessary credit.

"This portion of cellulose bound to lignin requires an additional delignification process before quantification in atmospheric PM, which requires harsh conditions and long reaction times (Gould, 1984; Kunit and Puxbaum, 1996)."

Line 117-123: "I have tried to find the "stark discrepancies" – in my opinion, this argument is a result of a sloppy reading past literature – by checking Sanchez-Ochoa, and Caseiro data I do not see such stark discrepancies and therefore recommend to include the larger past data sets into a comparison table or graph – and the differences will clear out. e.g.: the authors mixed up the absolute annual concentration trend – and the relative concentration related to OC. They neglected important characteristics of site differences – e.g. for the CARBOSOL project the differences in site characteristics, see May et al. Tellus 61B, 464-472, 2009. In fact, the overview of the present data, together with earlier published ones allows to offer an overview about communalities and differences, and increase the outcomes of scientific goals of the authors, to understand the atmospheric behavior and fate of plant debris."

This section has been revised, taking more care with the wording used. The section does not provide a direct comparison between the two studies but simply summarises each and suggests the need for more long-term characterisation studies. However, we still feel like the data provided by Caseiro (2008) does illustrate some differences from Sánchez-Ochoa et al. (2007) in cellulose concentrations. Even with comparison of only PM_{10} background sites from both studies, Caseiro (2008) show winter maxima at SCH and BB, compared to spring/summer at PDD and SIL from Sánchez-Ochoa et al. (2007). We appreciate the language used before was too simplified and reductionist, hence the revisions.

		Vienna			Styria			Salzburg			
		SCH	RIN	KEN	LOB	DB	GS	BB	RU	LE	AN
Cell.	avg	0.09	0.14	0.13	0.09	0.13	0.2	0.23	0.13	0.11	0.08
	min	0	0.03	0.04	0.02	0.04	0	0.11	0.05	0.03	0.04
	max	0.55	0.56	0.35	0.2	0.54	0.83	0.72	0.44	0.18	0.13
	winter	0.126	0.143	0.128	0.075	0.116	0.276	0.320	0.125	0.087	0.054
	spring	0.079	0.133	0.137	0.088	0.159	0.231	0.232	0.119	0.135	0.072
	summer	0.085	0.150	0.139	0.107	0.141	0.134	0.174	0.102	0.099	0.101
	autumn	0.083	0.120	0.098	0.079	0.114	0.156	0.200	0.163	0.090	0.074
Cell.	avg	1.57	1.37	1.6	1.4	1.91	1.48	1.4	1.17	1.62	1.68
-C	min	0.03	0.18	0.2	0.17	0.22	0	0.23	0.38	0.25	0.36
%	max	7.1	5.49	3.66	3.37	4.96	3.44	2.66	2.24	2.53	3.65
of	winter	1.660	1.040	1.168	0.906	0.929	0.798	0.990	0.757	0.905	0.950
OC	spring	1.373	1.288	1.844	1.402	2.590	1.986	1.470	1.107	2.004	1.867
	summer	1.813	1.987	2.342	1.970	2.273	1.710	1.943	1.214	1.831	2.489
	autumn	1.435	1.172	1.042	1.210	1.505	1.366	1.126	1.644	1.558	1.378



"Of the few previous characterisation studies to have taken place, only two have had a duration longer than one year. Regardless, some insights into the seasonal variations of cellulose concentrations have been afforded (Sánchez-Ochoa et al., 2007; Caseiro, 2008; Yttri et al., 2011a; Yttri et al., 2011b). For example, Sánchez-Ochoa et al. (2007) highlighted a pattern of cellulose concentration maxima during spring and summer at their rural background sites, excluding their maritime counterparts. This seasonal pattern, however, was found to be much weaker than other aerosol classes and showed higher winter concentrations than anticipated. Further, Caseiro (2008) found winter maxima at close to half their monitoring locations when observing from both urban and background locations. The reasons for the difference in seasonality between these two studies are likely to be owing to the differences in location and the variety of PM sizes used (PM_2 to PM_{10}) by Sánchez-Ochoa et al. (2007) compared to the consistent PM_{10} sampling used by Caseiro (2008). More long-term studies would be beneficial to understanding these geographical discrepancies."

Line 125-126: "Is here meant, that more data in addition to the current ones are needed, or is this an appraisal of the data given in this article?"

It is more meant as an appraisal of previous work, however this is not a suggestion that this dataset fulfils all needs regarding cellulose data collection.

Line 134-135: "I agree, that you have a very fine and for several reasons important data set. I recommend adding, that you have the chance for PM2.5/PM10 inter comparison, which is really a very important contribution, and the bi-annual data set (which had been also available in Carbosol, but with far lower time resolution)."

These comments have been added to the end of the introduction.

"Further, a $PM_{2.5}/PM_{10}$ intercomparison was also established"

Further Comments

"Why the intercomparison with earlier data is performed at the start of the chapter, and not at the end after showing your data?"

The intercomparison with earlier data was completed at the start of the chapter, since this is the first longer term cellulose study completed using the improved cellulose extraction protocol and the HPLC-PAD method, and so comparison with previous data to ensure our values were of the right order of magnitude was important.

"While I agree that local influences are actually very important, how can the relatively high absolute concentrations at the 3000 (SBO) and 1400 m (PDD) level can be explained?"

The relative high concentrations at both SBO and PDD are interesting indeed. Given the measurements at SBO were for $PM_{2.5}$, the long residence time of plant debris could lead to long distance travel and the eventual enrichment of plant debris at high altitudes. The supplementary data from the paper shows that seasonal average concentrations remain relatively consistent across the two years at SBO, so a consistent regional source (with wind speeds increasing with height – as the paper suggests) remains a possibility. One can assume that plant debris will reach the free troposphere after significant amount of time, during which significant abrasion/machining has taken place. PDD is surrounded by vegetation, compared to the sparse SBO site, which would suggest PDD is subject to local vegetation sources. The seasonal average concentrations over the two years show much greater variation in concentrations. It is possible that the 'local' biogenic sources surrounding PDD will vary more significantly than on a regional/continental scale for SBO.

The authors would also like to thank Prof Hans Puxbaum for their additional comments concerning cellulose sources. These comments have been taken into account and have been implemented into the introduction content where necessary (particularly line 97 – 126 of the revised ms).

"Plant debris (e.g. air-dispersed seeds or plant fragments via abrasion or decomposition mechanisms) is suspected to be a major contributor to PBAP within the atmosphere (Graham et al., 2003; Winiwarter et al., 2009; Martin et al., 2010; Yttri et al., 2011b; Bozzetti et al., 2016). However, atmospheric plant debris has received much less attention than other sources of PBAP, such as fungal spores, and thus knowledge of plant debris is severely limited. Both cellulose and plant waxes (as n-alkanes) have been used as proxy species for atmospheric plant debris. Early studies of the fraction of plant debris (or vegetative detritus) centred around analysis of plant waxes as the proxy species (Simoneit and Mazurek, 1982; Rogge et al., 1993a; Rogge et al., 1993b). These studies have formed the basis of our work, using identifiable chemical species to supply information on insoluble components. For example, Rogge et al. (1993a) in their experiment found significant amounts of non-extractable, insoluble organic components, yet were able to identify soluble components, such as plant waxes, as chemical tracers for insoluble components, such as plant debris. Rogge et al. (1993a) found local differences in the n-alkanes observed pattern, as a function of the variability in local plant composition, whilst Simoneit and Mazurek (1982) found plant wax to be a major component of rural OC."

"As scientific understanding increased, cellulose was proposed as a new chemical tracer for plant debris by Kunit and Puxbaum (1996) and has been used a tracer in several field and PMF studies since (Tenze-Kunit and Puxbaum, 2003; Sánchez-Ochoa et al., 2007; Caseiro, 2008; Yttri et al., 2011a,; Yttri et al., 2011b; Bozzetti et al., 2016; Borlaza et al., 2021a). Interestingly, Kotianová et al. (2008) evaluated the use of both plant waxes and cellulose as plant debris

tracers. They found a much weaker seasonal pattern with respect to cellulose concentrations, but showed plant wax/n-alkane concentrations peaked significantly during the warm summer months. The authors hypothesised that the difference between the two tracers revolved around plant waxes coming from the plant surface, whereas cellulose originating from bulk plant material. As such, atmospheric cellulose is predicted to be derived from machining and decomposition processes, and n-alkanes are emitted as part of surface abrasion mechanisms. Kotianová et al. (2008) found very good agreement in the results between the contributions of both cellulose and plant wax to PM_{10}."

Response to Anonymous Referee #2 (RC2)

The manuscript presents a comprehensive and valuable study on the atmospheric presence of cellulose based on an impressively large dataset. Cellulose is not the most critical species in atmospheric aerosol, yet a better understanding of its sources and atmospheric distribution is important in global aerosol and climate modelling. The work relies on the methodology of previous studies and in some respect contradicts to some of their findings. The sampling and analytical part of the study is scientifically sound, and the resulting dataset is reliable.

However, I have major reservations concerning emission source apportionment of cellulose based on correlation studies (sub-chapter 3.5). One of the findings of the authors is that biomass burning is not a source of cellulose for lack of correlation with the concentrations of the biomass burning tracer levoglucosan. *I never understand if there is a robust methodology for determining cellulose from particulates, and a hypothesis that biomass burning might release plant debris, why not test it by carrying out combustion experiments and determine cellulose from sampled smoke particulates? Why do the authors believe that simple correlation can prove or disprove such a hypothesis? Let us imagine a plausible scenario that is tested with correlation studies. Assume that cellulose are released from biomass burning, but not from wood burning where cellulose is strongly bound into the wood matrix, but from the burning of garden waste, of partially decomposed leaves and twigs, from which the escape of unburned plant debris may be possible. Beyond any doubts, atmospheric concentrations of levoglucosan will be governed by wood burning, since in any environment the mass of firewood dominates all the biomass that is burned. In this case, though cellulose IS released by a process of biomass burning, there will be no correlation between the measured concentrations of cellulose and levoglucosan simply because the dominant process (i.e. wood burning) overrides the signal. In addition, cellulose and levoglucosan should not come from the very same combustion process since levoglucosan is the pyrolysis product of cellulose itself. Thus, the plain statement that "the sources of atmospheric plant debris do not include any significant input from biomass burning" simply does not follow from the lack of correlation or negative correlation whatsoever.*

The same applies for EC correlated for proving resuspension sources of cellulose. Let resuspension be a source of atmospheric cellulose. Imagine that in a town it is raining for a week. Since there is traffic all week, EC concentrations will be measured but the concentration of cellulose will be zero simply because there is no resuspension under such circumstances. Can we conclude from the lack of correlation that resuspension is not a source of cellulose? Of course not. In addition, EC and cellulose are in different size ranges (fine vs coarse), are clearly from different sources (tailpipe vs resuspension), and are bound to different conditions (all weather vs. dry conditions). Why would anybody expect similar behaviour that

is manifested in perfect correlation? Simple analysis of resuspendable urban PM10 for cellulose would decide. Thus, the part of the manuscript on source apportionment by simple correlation is totally unfounded.

The authors would like to thank this referee for his contribution, and the good appraisal of our work in the first paragraph of his general comment.

We have to admit that it is really difficult to understand the point and the logical demonstration of the referee on how cellulose and levoglucosan could be emitted together in biomass combustion but not be correlated (the overall section we put in italic above). Maybe there is some kind of misunderstanding in the limits in our conclusions, and we made them clearer in the text and in the conclusions:

- We clearly mention at the beginning of the paper that we are essentially talking in this paper about free cellulose, and not cellulose still embedded in the lignin which is probably existing in the PM coming from plant debris. There is a possibility that this sort of cellulose could be emitted during biomass burning, and our conclusions are not addressing this point
- When talking about biomass burning we are only concerned with biomass burning from domestic wood, which is the very dominant process (as opposed to waste garden burning, which is prohibited in France) in the environments investigated.

In these conditions, we do not envisage processes removing cellulose from the atmosphere and leaving levoglucosan, if both are emitted at the same time from the same source, during full winter periods, and across multiple sampling campaigns, Further, regarding EC, our correlations are completed for datasets at least one year long, where meteorology changes consistently over time. Indeed, during rain there will be no resuspension of plant matter. However, EC concentrations would also be expected to be reduced due to wet deposition. Over a long time period and with a large array of data, we can produce a robust indication of co-emission.

"Thus, we can state that the sources of atmospheric plant debris, as indicated by measurements of free cellulose, do not seem to include any significant input from biomass burning from domestic wood. Further investigation would be needed concerning possible emissions of total cellulose (included the one still embedded in lignin."

Response to Anonymous Referee #3:

The manuscript by Brighty et al. describes long-term measurements of cellulose in atmospheric particles from nine sites across France and Switzerland. This is an important topic since the contribution of cellulose to organic carbon is not negligible, particularly in rural areas, and because current information about the sources and atmospheric distribution of this polysaccharide is very scarce.

The manuscript has no major problems. It is well written and is easy to follow. The research seems to be well-planned and conducted. The obtained results make sense, and are clearly presented and discussed. The conclusions are supported by the results.

I believe that this study will be of great value to the community of atmospheric chemistry researchers and recommend publication in ACP after the following minor comments are addressed:

The authors would like to thank this referee for his nice comment about our work, and his contribution in helping to refine and polish this report.

Title: "The title should be more informative. I suggest something like "Cellulose in atmospheric particulate matter at rural and urban sites across central Europe"."

Corrected to be more informative, as suggested.

"Cellulose in atmospheric particulate matter at rural and urban sites in Europe"

Lines 324 to 333: "Supplementary information should not be essential to understand the comparison with previous data. Therefore, tick labels in Figure 2 should include the countries names after the sampling site names (or acronyms). If not possible, because of the length of tick labels, this information should be added to the figure caption."

Countries of the sampling sites have been added to the figure caption as follow :

Taken from the paper -->

Countries for literature sampling sites:

PM_{10} Urban: *ROT – Netherlands; Oslo – Norway; RIN, KEN, DB, GS, RU and LE – Austria.*

PM_{10} Rural: *SIL – Germany; PDD – France; BIR, Hurdal and Hyyttiälä – Norway; Lille Valby, VAV – Denmark; SCH, LOB, BB and AN – Austria.*

$PM_{2.5}$ or smaller: *AZO, AVE – Portugal; KPZ – Hungary; SBO – Austria; Oslo, Hurdal – Norway.*

Line 364: "At the ANDRA-OPE sampling site, the contribution of cellulose in PM2.5 to that in PM10 seems to be much lower than 18%. Please, check if there is a calculation error here."

There is no calculation error, but there were simply not many $PM_{2.5}$ measurements being taken across the year and PM_{10} concentrations did fluctuate significantly across the year.

Table 4: "Sampling site name should be corrected to ANDRA-OPE."

Corrected to ANDRA-OPE

Line 372: "The authors should provide an explanation for not having calculated the PM2.5/PM10 ratio at the ANDRA-OPE site. Do you have any explanation for the low levels of cellulose in PM2.5 at this site?"

The ANDRA-OPE data for $PM_{2.5}$ is only from 2020, which we suspect has affected cellulose emission sources due to the COVID-19 pandemic. This is spoken about later in section 3.7. The explanation for the lack of $PM_{2.5}/PM_{10}$ ratio is explained now in the text.

"The comparatively low cellulose concentrations at ANDRA-OPE for 2020 (both PM_{10} and $PM_{2.5}$) are discussed as part of section 3.7, in the interannual comparison. No ratio is provided at ANDRA-OPE as $PM_{2.5}$ and PM_{10} measurements were completed on different days, as opposed to simultaneous PM_{10} and $PM_{2.5}$ sampling at Payerne and Zurich."

Lines 374-375: "References to previous studies reporting a higher abundance of cellulose in the coarse mode should be given."

Suitable references added for previous studies.

"This large data set of size resolved cellulose concentrations confirms that plant debris predominantly resides within the coarse aerosol mode (Sánchez-Ochoa et al., 2007; Yttri et al., 2011a)"

Lines 390-391: "The relationship between biological activity and meteorological conditions needs to be better explained. A higher abundance of plant debris from decaying leaves is expected to occur in autumn. How is it related with the summer to autumn temperature decrease and humidity increase?"

The relationship between meteorological conditions and seasonality has been explored further in the report.

"In general, the seasonal pattern exhibited here shows higher cellulose concentrations during summer and autumn, likely due to increased temperature and humidity increasing the activity of soil and litter decomposers as well as improving the quality of the litter composition. For example, nitrogen content of leaves is shown to be greater in warmer temperatures, which leads to better conditions for leaf degradation by microbial action (Liu et al., 2006; Verma et al., 2018). This hypothesis would require further experiments, including specific field measurements linking soil and litter state and plant debris emission"

Line 632: "Table sequence needs to be changed in the supplementary material file in order to follow the same sequence of the Results and Discussion section."

Supplementary material figures and tables have been reordered correctly.

Table S1: "Table sequence needs to be changed in the supplementary material file in order to follow the same sequence of the Results and Discussion section."

Site typology has been added to the table, as suggested.