Manuscript acp-2020-1065 entitled "Determination of free amino acids, saccharides and selected microbes in biogenic atmospheric aerosols - seasonal variations, particle size distribution, chemical and microbial relations"

Editor Decision: Reject (12 Feb 2021) by Aurélien Dommergue
Comments to the Author:
Dear Author,

I am sorry to inform you that after careful discussions with the reviewers I have decided to reject your manuscript.
The reviewer 2 raise important concerns about some results and more importantly underlined that both chemical data and gene copies data have been modified. This appears to question the validity of the study, and it is very suspicious that in the revised manuscript version, values in Tables and Figures were changed without any explanation. The statement "The data in all Tables and in all Figures have been checked, revised and updated " does not suffice to provide a thorough documentation why the values differ among the manuscript versions that will be all publicly accessible.

Given that the revision did not lead to a sufficiently improved manuscript and even more so, the data is doubtful, I have decided to reject this manuscript.

Sincerely yours
AD

Authors' response to Editor: Original data matrix, including actual concentrations of the chemical compounds and DNA and gene copies numbers of microbes, was transformed, using logarithmic transformation, to achieve normal data distribution as described in the text. This was compulsory for the adequate interpretation of the different statistical algorithms used in the manuscript. The data processing was successfully developed using the normal distributed data. This data (logarithmic transformed) was originally used for the development of Figures 1 and 2 and Tables S8-11. During original manuscript preparation for the clarification we decided to use the actual concentrations of the chemical compounds and the total DNA and the gene copy numbers of the microorganisms in these figures and tables, instead of the logarithmic transformed values. Then the actual concentrations and gene copy numbers without outliers were calculated from the normalized data. During this process an unidentified error, which affected the concentrations and the gene copy numbers, showed in figures and tables of the original manuscript, was encounted. This error was only detected thanks to Referee #1 during the reviewing process of Discussion manuscript version. We followed the valuable comments of Referee #1 and calculations based on the correct data plotted in Figure 1 were used for Figure 2 and Tables S8-11 of the revised MS version. We are very sorry, but unfortunately by our mistake, uncorrected Figures 3 and S6 were included in the revised version of the manuscript (corrected Figure versions in the end of this letter).However, all the calculations described in the manuscript were done with the logarithmic dataset taken from the first Discussion version, and the revision of values of Figures 1 and 2, and Tables S8-11 did not affect these calculations, nor discussions.

Answers to the comments of referees (Report #1 and Report #2)

Report #1

Submitted on 20 Jan 2021

Referee #2: Romie Tignat-Perrier, rom26.p@hotmail.fr

Recommendation to the editor

1) Scientific significance Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?	Outstanding Excellent **Good** Fair Low
2) Scientific quality Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?	Outstanding **Excellent** Good Fair Low
3) Presentation quality Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?	Outstanding Excellent **Good** Fair Low

For final publication, the manuscript should be

accepted as is

accepted subject to **technical corrections**

accepted subject to **minor revisions**

reconsidered after **major revisions**

I would be willing to review the revised paper, if the editor considers it necessary.

I am **not** willing to review the revised paper.

rejected

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

Report #2

Submitted on 02 Feb 2021

Referee #1: Pierre Amato, pierre.amato@uca.fr

Recommendation to the editor

1) Scientific significance Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?	Outstanding Excellent Good **Fair** Low
2) Scientific quality Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?	Outstanding Excellent Good **Fair** Low
3) Presentation quality Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?	Outstanding Excellent Good **Fair** Low

For final publication, the manuscript should be

accepted as is

accepted subject to **technical corrections**

accepted subject to **minor revisions**

reconsidered after **major revisions**

I would be willing to review the revised paper, if the editor considers it necessary.

I am **not** willing to review the revised paper.

rejected

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

Comments posted as supplement.

Referee Report: acp-2020-1065-referee-report.pdf

Comments on Ruiz-Jimenez et al: «Chemical and microbiological characterization of primary biological aerosol particles at the boreal forest»revised into “Determination of free amino acids, saccharides and selectedmicrobes in biogenic atmospheric aerosols -seasonal variations,particle size distribution, chemical and microbial relations”I have to admit that I am a bit confused by the revision and concerned by the validity of the data and analyses. I do not follow the argument of the samples beingcollected in September-November in the present study and that would explain the low values of gene copy numbers. On the contrary the values should be much higher than those reported, referring to fig2 in Helin et al. (~1000 genes copies/m3 at least as expressed as bacteria/m3 in Helin).In addition, I do not really get the point regarding the fraction of filter used for

explaining the low values, since the data were normalized to air volumes (was normalization to air volume somehow not linearly related to filter surface??).

References: Helin, A., Sietiö, O.-M., Heinonsalo, J., Bäck, J., Riekkola, M.-L. and Parshintsev, J.: Characterization of free amino acids, bacteria and fungi in size-segregated atmospheric aerosols in boreal forest: seasonal patterns, abundances and size distributions, Atmospheric Chemistry and Physics, 17(21), 13089–13101, https://doi.org/10.5194/acp-17-13089-2017, 2017.

Authors' response: The reason for the differences in gene copy numbers between 2014 and 2017 can be due to two aspects:
1)Climatic conditions/meteorological variables. As can be seen from Table below, the total number of particles in 2017 was around a half of that in 2014 supporting the difference between the results for genes copies/m3. Namely in Finland the annual variation in air microbial concentrations can be very high due to large differences in seasonal weather conditions.

Year	Month	T air (degC)	Precipitation (mm)	UV a (W m-2)	UV b (W m-2)	T soil (degC)	GPP (µmol m-2 s-1)	TNP*
2014	Sep	10.1	0.7	5.7	0.3	10.7	4.5	2632.1
	Oct	4.1	1.8	1.9	0.1	6.0	1.5	1515.1
2017	Sep	9.2	2.1	4.5	0.2	10.0	4.3	1334.9
	Oct	3.6	3.4	1.6	0.1	6.0	1.4	973.5

*TNP = total number of particles

2) Technical limitations regarding to the assay itself due to lower sample amount. We wanted to make chemical and DNA analysis from the same sample and that's why we divided the filter into two equal pieces, one for both analysis. This might give higher detection level as previously, but not affect the gene copy numbers/m3.

The data in all Tables and in all Figures have been checked, revised and updated". I am very puzzled here: all or almost all the values for amino acids, saccharides and gene copies have changed (increased, by different factors) in Figures 1, 2, Tables S8-S11 compared to the last version, whereas the data were not changed in other figures (Figure 3 and Figure S6 for example). The modifications done would need to be at least clearly listed and justified.

Authors' response: Original data matrix, including actual concentrations of the chemical compounds and DNA and gene copies numbers of microbes, was transformed, using logarithmic transformation, to achieve normal data distribution as described in the text. This was compulsory for the adequate interpretation of the different statistical algorithms used in the manuscript. The data processing was successfully developed using the normal distributed data. This data (logarithmic transformed) was originally used for the development of Figures 1 and 2 and Tables S8-11. During original manuscript preparation for the clarification we decided to use the actual concentrations of the chemical compounds and the total DNA and the gene copy numbers of the microorganisms in these figures and tables, instead of the logarithmic transformed values. Then the actual concentrations and gene copy numbers without outliers were calculated from the normalized data. During this process an unidentified error, which affected the concentrations and the gene copy numbers, showed in figures and tables of the original manuscript, was encounted. This error was only detected thanks to Referee #1 during the reviewing process of Discussion manuscript version. We followed the valuable comments of Referee #1 and calculations based on the correct data plotted in Figure 1 were used for Figure 2 and Tables S8-11 of the revised MS version. We are very sorry, but unfortunately by our mistake, uncorrected Figures 3 and S6 were included in the revised version of the manuscript (corrected Figure versions in the end of this letter). However, all the calculations described in the manuscript were done with the logarithmic

dataset taken from the first Discussion version, and the revision of values of Figures 1 and 2, and Tables S8-11 did not affect these calculations, nor discussions.

Averages, standard deviationsand CVs, min/max and ranges were modified (Tables S8-S11), but surprisingly neither skewness nor kurtosis were affected.

Authors' response: Skewness and kurtosis test were developed using the logarithmic transformed data (this transformation was done to ensure the normal distribution of the data used in the statistical analysis), as described in the Table legends. No errors were found in the logarithmic data, therefore the values were not affected.

I would then also expect the regression coefficients (based on parametric statistics) to change according to raw data modification, since the modifications of the data were not linear, but this is surprisingly not the case (Fig 4 and 5,S4,S7,etc). For example: bacteria in PM2.5-10 (Table S10) were modified from 4.4 +/-1.7 (average +/-SD) into 53.0 +/-54.0(factor of 12 between averages). Elsewhere (Table S8) Gln in PM2.5-10 was modified from 1.9 +/-1.9over into 5.6 +/-4.7(factor of ~3 between averages). However, the Pearson correlation between Bact and Gln is still the same as in the previous version (Figure S4).

Authors' response: All the calculations described in the original and revised manuscripts were done with the logarithmic dataset taken from the first Discussion version, and the revision of values of Figures 1 and 2, and Tables S8-11 did not affect these calculations, nor discussions in the revised manuscript.

Figure 3

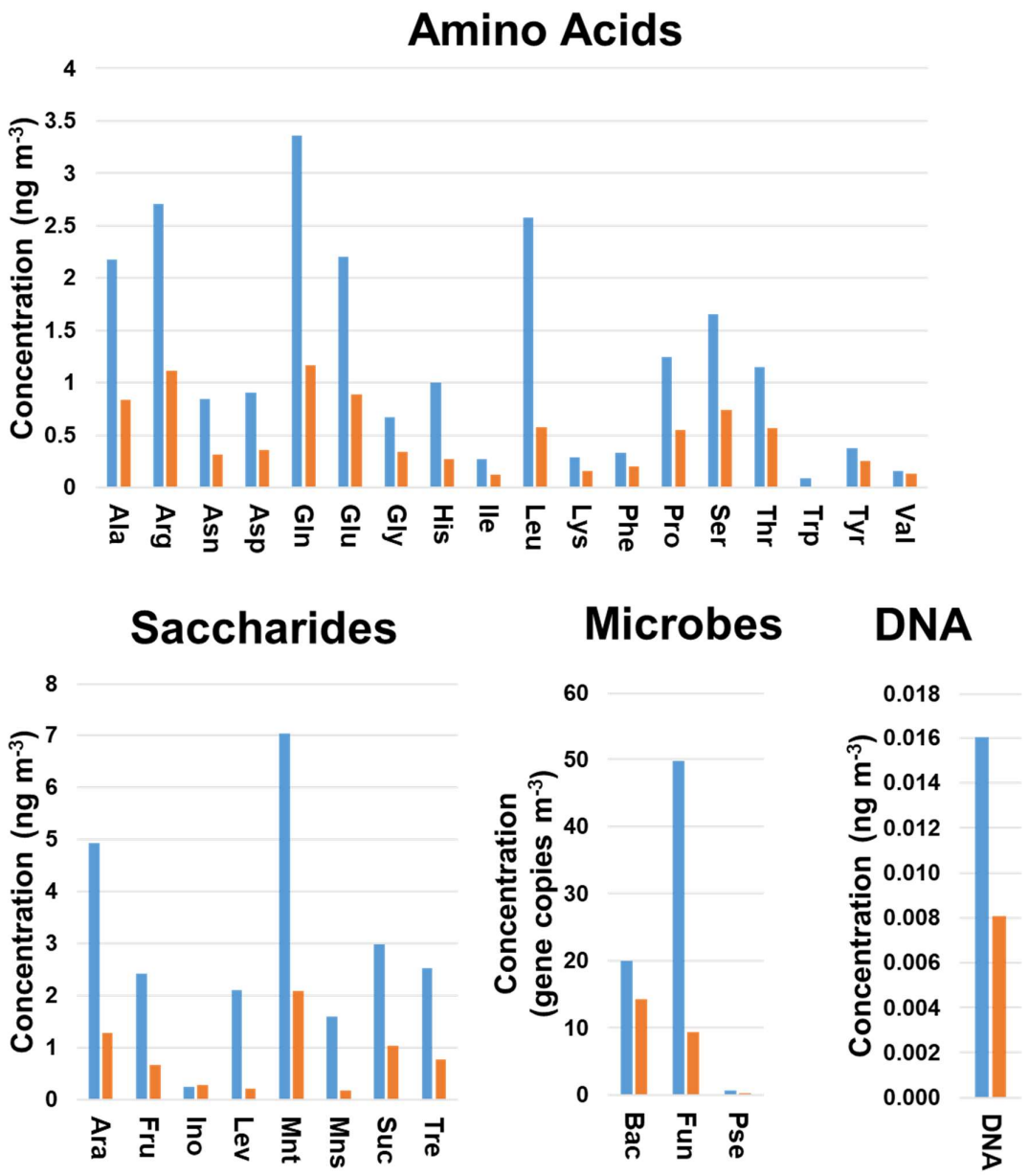
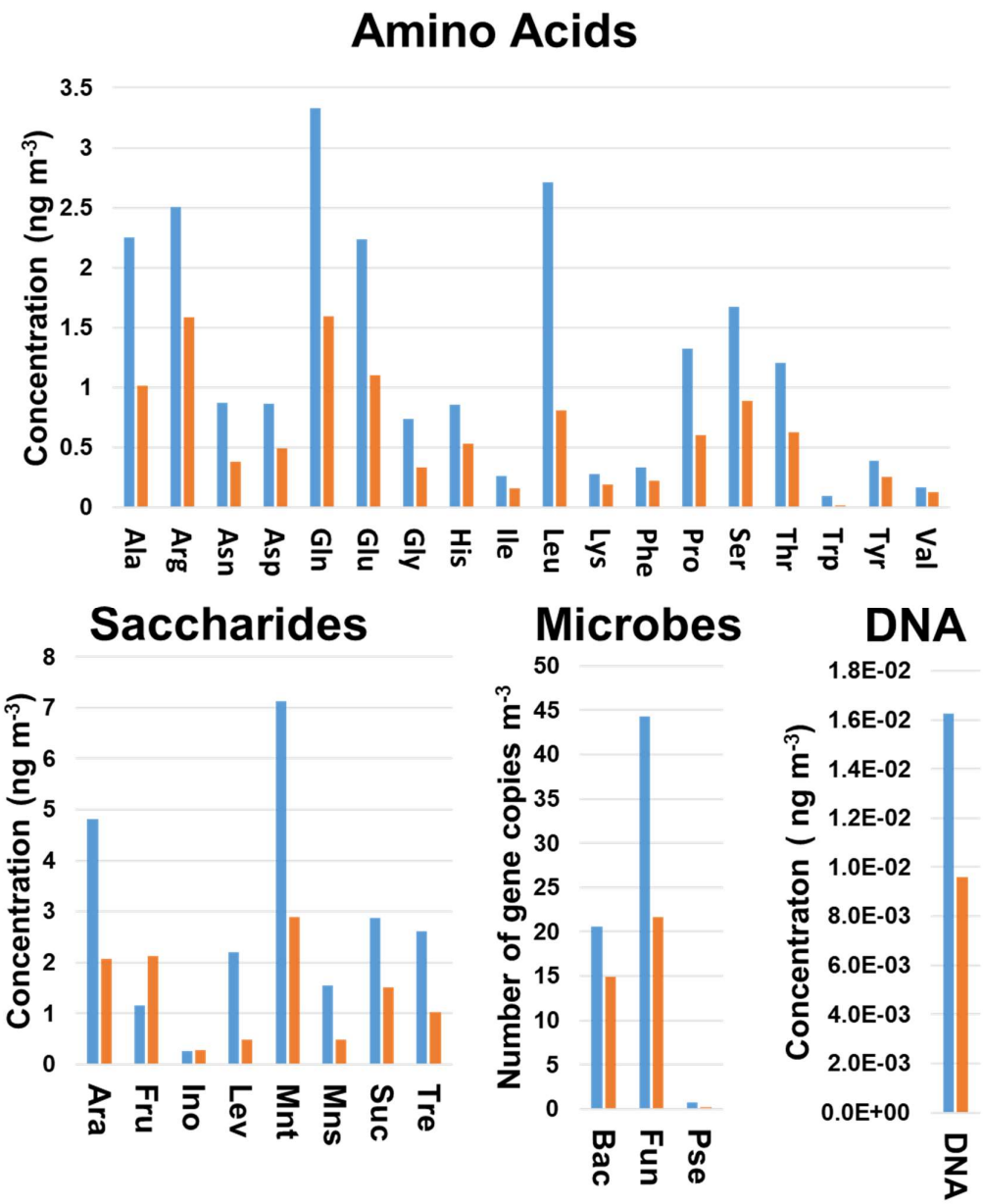

Amino Acids
Concentration (ng m-3)
4
3.5
3
2.5
2
1.5
1
0.5
0
Ala
Arg
Asn
Asp
Gln
Glu
Gly
His
Ile
Leu
Lys
Phe
Pro
Ser
Thr
Trp
Tyr
Val
Saccharides
Concentration (ng m-3)
8
7
6
5
4
3
2
1
0
Ara
Fru
Ino
Lev
Mnt
Mns
Suc
Tre
Microbes
Concentration (gene copies m-3)
60
50
40
30
20
10
0
Bac
Fun
Pse
DNA
Concentration (ng m-3)
0.018
0.016
0.014
0.012
0.010
0.008
0.006
0.004
0.002
0.000
DNA

Figure S6 (B)


Amino Acids
Concentration (ng m-3)
3.5
3
2.5
2
1.5
1
0.5
0
Ala
Arg
Asn
Asp
Gln
Glu
Gly
His
Ile
Leu
Lys
Phe
Pro
Ser
Thr
Trp
Tyr
Val
Saccharides
Concentration (ng m-3)
8
7
6
5
4
3
2
1
0
Ara
Fru
Ino
Lev
Mnt
Mns
Suc
Tre
Microbes
Number of gene copies m-3
50
45
40
35
30
25
20
15
10
5
0
Bac
Fun
Pse
DNA
Concentraton (ng m-3)
1.8E-02
1.6E-02
1.4E-02
1.2E-02
1.0E-02
8.0E-03
6.0E-03
4.0E-03
2.0E-03
0.0E+00
DNA