Reviewer 2

General Comments

The purpose of this paper is to describe the molecular composition of dissolved organic matter in cloud water at a novel site in Reunion Island and compare it to Puy de Dome in France using primarily FT-ICR MS analysis. The samples are also compared to previous studies and use various metrics to evaluate the composition of the samples for comparison.

Overall, I feel this is a good paper that lays good groundwork for the analysis of cloud water in remote areas that have not previously been investigated with this type of analysis. There are some things that I am interested in and things that should be addressed before full publication, but they are relatively minor and should not hinder its publication in my view.

We would like to acknowledge the reviewer for the accurate for her/his constructive comments and suggestions. The concerns raised helped us to look more in deep in the data and to improve the quality and the presentation of the results.

Specific Comments

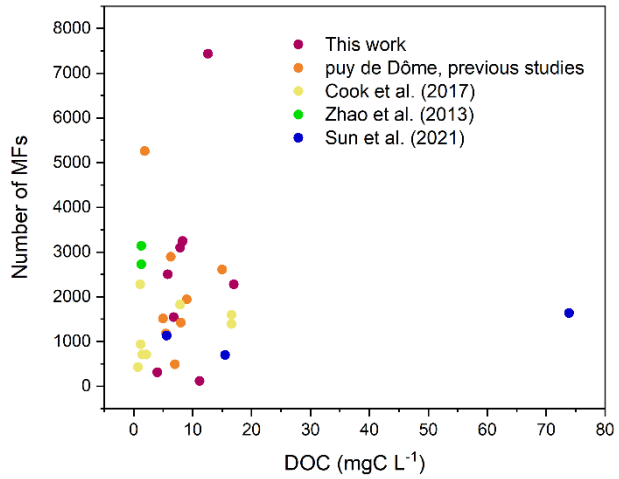
Line 177: MFAssignR also incorporates H2O, CH2O, and O homologous series for formula extension. A citation of the package on GitHub, or the manuscript itself (Schum et al. Env. Res. 2020) would be a good addition to this section as well.

We agree with the reviewer. The reference was added in this section at line 168.

Line 274-275: Is there an explanation for why 22/10/2019 has so few MF compared to 8/10/2019? Or maybe why 8/10/2021 has so many more than the rest of the samples? It seems like the DOC is pretty similar between them, with the main differences coming from the inorganic ions. Do you think it is related to the actual sample itself, or to the blank subtraction method? Conservative blank subtraction is a good choice, but I am curious what the formula numbers looked like prior to blank subtraction and whether they were more similar at that point.

We are persuaded that the very variable number of MFs is related to the sample itself, as shown in the following table that reports the number of MFs before and after blank subtraction for each sample presented in this work. We already observed very different numbers of MFs in cloud samples in Bianco et al., (2019) and in Cook et al., (2017). The following plot represents the DOC vs ESI FT-ICR MS number of MF for samples from the PUY and the REU. Samples presented in Bianco et al., (2018) and 2019 (9.4 T, Bruker Solarix), Cook et al., 2017 (12T Bruker SolariX), Zhao et al., (2013) (LTQFTUltra, ThermoScientific), and Sun et al., (2021) (9.4-T Bruker SolariX XR) are presented in the plot. It's worth noting that no trend is observed between the DOC concentration and the number of MFs in the ESI FT-ICR MS response. As noted before, samples from the PUY show a huge variability in the number of MFs. Moreover, the work of Cook et al. 2017 also shows a huge variability in the number of MF for samples in the same range of DOC concentration (between 430 and 2300 MF).

Sample	#MF before blank subtraction	#MF after blank subtraction	#MF in common
R8	3199	3098	101
R9	2739	2503	236
R10B	2463	2276	187
02/03/2019	3441	3244	197
15/03/2019	2345	2084	261
02/10/2019	1555	1543	12
22/10/2019	222	120	102
17/07/2020	849	715	134
03/11/2020	433	312	121
08/10/2021	8073	7436	637



This interesting result is now reported in the text.

Lines 285-288: "This variability is due to the influence of primary emissions and to the atmospheric reactivity: a similar variability in the number of MFs has been already observed in previous works (Zhao et al., 2013; Cook et al., 2017; Sun et al., 2021; Bianco et. al., 2018; Bianco et al., 2019). In addition, the DOC concentration and the number of MFs show no correlation in this work as well as in published data (Figure S1)."

Lines 303-304: You mention that the average OSC is similar between PUY and REU autumn samples, while this can definitely just be a coincidence (considering the different sources and conditions) I was curious if you looked into the molecular formulas to see what sort of differences occurred in them. For example is the OSC heavily influenced in both cases by a common set of molecular formulas (even if they are different molecules) or are there really no similarities at all, they just happen to average out to the same OSC?

We agree with the reviewer that this point is important and needs more discussion. Nevertheless, it is not easy to find an approach to answer this question.

We started by comparing the molecular formulas in the presented samples through a Venn diagram, which is difficult to represent for the whole dataset. The following table present the different intersections for the Venn diagram and the number of MF for each intersection. Surprisingly, only 8 MFs are common to all

the samples (line 1, in yellow). These formula are CHO compounds, namely $C_{15}H_{26}O_8$, $C_{14}H_{22}O_9$, $C_{11}H_{12}O_4$, $C_{17}H_{26}O_9$, $C_{10}H_{14}O_6$, $C_9H_{16}O_6$, $C_{15}H_{24}O_9$, and $C_7H_8O_5$, with OSC values between -0.18 and +0.77. The number of MFs contained in 9 samples is 46, in 8 samples is 108, in 7 samples is 250, in 6 samples is 586. MFs contained only in one sample are reported in the last ten lines of the table and represent from 5 to 50% of the total number of MFs in the sample.

Intersection between										#MF
02/03/2019	***02/10/2019***	***03/11/2020***	***08/10/2021***	***15/03/2019***	***17/07/2020***	***22/10/2019***	***R10B***	***R8***	***R9***	***8***
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	22/10/2019	R10B	R8	R9		9
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	R10B	R8	R9		35
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	22/10/2019	R8	R9		2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	22/10/2019	R10B	R8	R9			6
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	R10B	R8	R9			77
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	R10B	R8	R9			18
02/03/2019	02/10/2019	08/10/2021	17/07/2020	22/10/2019	R10B	R8	R9			1
02/03/2019	02/10/2019	03/11/2020	08/10/2021	17/07/2020	R10B	R8	R9			1
02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	R10B	R8	R9			3
02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	22/10/2019	R8	R9			1
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	R10B	R9			1
02/03/2019	02/10/2019	08/10/2021	15/03/2019	R10B	R8	R9				162
02/03/2019	08/10/2021	15/03/2019	22/10/2019	R10B	R8	R9				3
02/03/2019	03/11/2020	15/03/2019	17/07/2020	R10B	R8	R9				1
02/03/2019	08/10/2021	15/03/2019	17/07/2020	R10B	R8	R9				6
02/03/2019	03/11/2020	08/10/2021	15/03/2019	R10B	R8	R9				7
02/03/2019	02/10/2019	08/10/2021	17/07/2020	R10B	R8	R9				7
02/03/2019	02/10/2019	03/11/2020	08/10/2021	R10B	R8	R9				1
02/10/2019	08/10/2021	15/03/2019	17/07/2020	R10B	R8	R9				6
02/10/2019	03/11/2020	08/10/2021	17/07/2020	R10B	R8	R9				6
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	R8	R9				2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	R10B	R8				1
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	R10B	R8				1
02/03/2019	02/10/2019	03/11/2020	08/10/2021	17/07/2020	R10B	R8				1
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	R8				2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	22/10/2019	R10B	R9				1
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	R10B	R9				2
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	R9				1
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	22/10/2019				40
02/03/2019	02/10/2019	15/03/2019	R10B	R8	R9					5
02/03/2019	08/10/2021	15/03/2019	R10B	R8	R9					416
02/03/2019	02/10/2019	08/10/2021	R10B	R8	R9					23

Intersection between										#MF
02/10/2019	08/10/2021	15/03/2019	R10B	R8	R9					7
08/10/2021	15/03/2019	17/07/2020	R10B	R8	R9					2
02/10/2019	08/10/2021	22/10/2019	R10B	R8	R9					1
02/10/2019	03/11/2020	17/07/2020	R10B	R8	R9					3
02/10/2019	08/10/2021	17/07/2020	R10B	R8	R9					41
02/10/2019	03/11/2020	08/10/2021	R10B	R8	R9					2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	R8	R9					7
02/03/2019	02/10/2019	08/10/2021	17/07/2020	R8	R9					1
02/10/2019	03/11/2020	08/10/2021	17/07/2020	R8	R9					2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	R10B	R8					7
02/03/2019	02/10/2019	08/10/2021	17/07/2020	R10B	R8					1
02/10/2019	03/11/2020	08/10/2021	17/07/2020	R10B	R8					3
02/03/2019	02/10/2019	08/10/2021	15/03/2019	22/10/2019	R8					1
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	R8					1
02/03/2019	02/10/2019	03/11/2020	08/10/2021	17/07/2020	R8					1
02/03/2019	02/10/2019	08/10/2021	15/03/2019	R10B	R9					13
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	R10B					1
02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	R10B					1
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020	22/10/2019					6
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	22/10/2019					4
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020					36
02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020	22/10/2019					1
02/03/2019	15/03/2019	R10B	R8	R9						71
02/03/2019	02/10/2019	R10B	R8	R9						1
02/03/2019	08/10/2021	R10B	R8	R9						124
08/10/2021	15/03/2019	R10B	R8	R9						21
02/10/2019	17/07/2020	R10B	R8	R9						1
02/10/2019	08/10/2021	R10B	R8	R9						39
08/10/2021	17/07/2020	R10B	R8	R9						8
02/03/2019	08/10/2021	15/03/2019	R8	R9						75
02/03/2019	02/10/2019	08/10/2021	R8	R9						9
02/10/2019	08/10/2021	15/03/2019	R8	R9						4
02/10/2019	03/11/2020	17/07/2020	R8	R9						1
02/10/2019	08/10/2021	17/07/2020	R8	R9						6
02/03/2019	15/03/2019	17/07/2020	R10B	R8						1
02/03/2019	08/10/2021	15/03/2019	R10B	R8						35
02/03/2019	02/10/2019	08/10/2021	R10B	R8						1
02/10/2019	08/10/2021	17/07/2020	R10B	R8						6
03/11/2020	08/10/2021	17/07/2020	R10B	R8						1

Intersection between										#MF
02/03/2019	02/10/2019	08/10/2021	15/03/2019	R8						8
02/03/2019	03/11/2020	08/10/2021	15/03/2019	R8						1
02/10/2019	03/11/2020	08/10/2021	17/07/2020	R8						1
02/03/2019	02/10/2019	15/03/2019	R10B	R9						1
02/03/2019	08/10/2021	15/03/2019	R10B	R9						28
02/03/2019	02/10/2019	08/10/2021	R10B	R9						1
02/10/2019	08/10/2021	15/03/2019	R10B	R9						1
02/10/2019	08/10/2021	17/07/2020	R10B	R9						2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	R9						2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	R10B						2
02/03/2019	02/10/2019	08/10/2021	17/07/2020	R10B						1
02/10/2019	03/11/2020	08/10/2021	17/07/2020	R10B						2
02/03/2019	02/10/2019	08/10/2021	15/03/2019	22/10/2019						1
02/03/2019	02/10/2019	03/11/2020	15/03/2019	17/07/2020						1
02/03/2019	02/10/2019	08/10/2021	15/03/2019	17/07/2020						30
02/03/2019	02/10/2019	03/11/2020	08/10/2021	15/03/2019						13
02/03/2019	02/10/2019	03/11/2020	08/10/2021	17/07/2020						1
02/10/2019	03/11/2020	15/03/2019	17/07/2020	22/10/2019						1
02/10/2019	03/11/2020	08/10/2021	15/03/2019	17/07/2020						6
02/10/2019	03/11/2020	08/10/2021	17/07/2020	22/10/2019						6
02/03/2019	R10B	R8	R9							64
15/03/2019	R10B	R8	R9							15
02/10/2019	R10B	R8	R9							50
03/11/2020	R10B	R8	R9							1
08/10/2021	R10B	R8	R9							135
02/03/2019	15/03/2019	R8	R9							7
02/03/2019	02/10/2019	R8	R9							1
02/03/2019	08/10/2021	R8	R9							71
08/10/2021	15/03/2019	R8	R9							7
02/10/2019	08/10/2021	R8	R9							24
08/10/2021	17/07/2020	R8	R9							1
02/03/2019	15/03/2019	R10B	R8							4
02/03/2019	08/10/2021	R10B	R8							14
08/10/2021	15/03/2019	R10B	R8							5
02/10/2019	08/10/2021	R10B	R8							4
08/10/2021	17/07/2020	R10B	R8							3
02/03/2019	02/10/2019	15/03/2019	R8							2
02/03/2019	08/10/2021	15/03/2019	R8							53
02/03/2019	02/10/2019	08/10/2021	R8							4

Intersection between										#MF
02/03/2019	03/11/2020	08/10/2021	R8							1
02/10/2019	08/10/2021	15/03/2019	R8							1
02/10/2019	08/10/2021	22/10/2019	R8							2
02/10/2019	03/11/2020	17/07/2020	R8							1
02/10/2019	08/10/2021	17/07/2020	R8							10
03/11/2020	08/10/2021	17/07/2020	R8							2
02/03/2019	15/03/2019	R10B	R9							20
02/03/2019	02/10/2019	R10B	R9							2
02/03/2019	08/10/2021	R10B	R9							24
08/10/2021	15/03/2019	R10B	R9							2
02/10/2019	08/10/2021	R10B	R9							2
02/03/2019	08/10/2021	15/03/2019	R9							23
02/03/2019	02/10/2019	08/10/2021	R9							2
02/10/2019	03/11/2020	17/07/2020	R9							1
02/03/2019	08/10/2021	15/03/2019	R10B							16
02/10/2019	08/10/2021	17/07/2020	R10B							2
02/03/2019	02/10/2019	15/03/2019	17/07/2020							2
02/03/2019	02/10/2019	08/10/2021	15/03/2019							60
02/03/2019	08/10/2021	15/03/2019	22/10/2019							1
02/03/2019	08/10/2021	15/03/2019	17/07/2020							6
02/03/2019	03/11/2020	08/10/2021	15/03/2019							10
02/03/2019	02/10/2019	17/07/2020	22/10/2019							1
02/03/2019	02/10/2019	03/11/2020	17/07/2020							1
02/03/2019	02/10/2019	08/10/2021	17/07/2020							2
02/03/2019	03/11/2020	08/10/2021	17/07/2020							1
02/10/2019	08/10/2021	15/03/2019	17/07/2020							2
02/10/2019	03/11/2020	17/07/2020	22/10/2019							3
02/10/2019	08/10/2021	17/07/2020	22/10/2019							1
02/10/2019	03/11/2020	08/10/2021	17/07/2020							32
R10B	R8	R9								123
02/03/2019	R8	R9								45
15/03/2019	R8	R9								5
02/10/2019	R8	R9								5
08/10/2021	R8	R9								147
02/03/2019	R10B	R8								5
15/03/2019	R10B	R8								3
17/07/2020	R10B	R8								1
08/10/2021	R10B	R8								52
02/03/2019	15/03/2019	R8								10

Intersection between										#MF
02/03/2019	08/10/2021	R8								65
08/10/2021	15/03/2019	R8								18
02/10/2019	17/07/2020	R8								5
02/10/2019	08/10/2021	R8								57
08/10/2021	17/07/2020	R8								6
03/11/2020	08/10/2021	R8								1
02/03/2019	R10B	R9								44
15/03/2019	R10B	R9								2
02/10/2019	R10B	R9								3
17/07/2020	R10B	R9								1
08/10/2021	R10B	R9								24
02/03/2019	15/03/2019	R9								9
02/03/2019	02/10/2019	R9								1
02/03/2019	08/10/2021	R9								21
08/10/2021	15/03/2019	R9								6
02/10/2019	08/10/2021	R9								3
08/10/2021	17/07/2020	R9								1
02/03/2019	15/03/2019	R10B								9
02/03/2019	02/10/2019	R10B								2
02/03/2019	08/10/2021	R10B								10
08/10/2021	15/03/2019	R10B								5
02/10/2019	08/10/2021	R10B								4
02/03/2019	02/10/2019	15/03/2019								3
02/03/2019	03/11/2020	15/03/2019								2
02/03/2019	08/10/2021	15/03/2019								203
02/03/2019	02/10/2019	08/10/2021								6
02/03/2019	03/11/2020	17/07/2020								1
02/03/2019	08/10/2021	17/07/2020								2
02/10/2019	03/11/2020	15/03/2019								1
02/10/2019	08/10/2021	15/03/2019								9
08/10/2021	15/03/2019	17/07/2020								1
03/11/2020	08/10/2021	15/03/2019								1
02/10/2019	17/07/2020	22/10/2019								7
02/10/2019	03/11/2020	22/10/2019								1
02/10/2019	08/10/2021	22/10/2019								2
02/10/2019	03/11/2020	17/07/2020								11
02/10/2019	08/10/2021	17/07/2020								51
02/10/2019	03/11/2020	08/10/2021								1
03/11/2020	08/10/2021	17/07/2020								3

Intersection between										#MF
R8	R9									68
R10B	R8									49
02/03/2019	R8									31
15/03/2019	R8									9
02/10/2019	R8									2
17/07/2020	R8									1
08/10/2021	R8									370
R10B	R9									70
02/03/2019	R9									26
15/03/2019	R9									4
17/07/2020	R9									1
08/10/2021	R9									34
02/03/2019	R10B									27
15/03/2019	R10B									9
02/10/2019	R10B									2
08/10/2021	R10B									22
02/03/2019	15/03/2019									131
02/03/2019	02/10/2019									1
02/03/2019	17/07/2020									1
02/03/2019	08/10/2021									245
02/10/2019	15/03/2019									2
08/10/2021	15/03/2019									67
02/10/2019	22/10/2019									1
02/10/2019	17/07/2020									57
02/10/2019	03/11/2020									2
02/10/2019	08/10/2021									91
03/11/2020	22/10/2019									1
08/10/2021	22/10/2019									1
03/11/2020	17/07/2020									2
08/10/2021	17/07/2020									26
03/11/2020	08/10/2021									5
R8										***233***
R9										***123***
R10B										***209***
02/03/2019										***626***
15/03/2019										***134***
02/10/2019										***290***
22/10/2019										***7***
17/07/2020										***78***

Intersection between										#MF
03/11/2020										***11***
08/10/2021										***3929***

We also calculated the intersections of the lists of MFs, which are presented in the following table, where samples are reported in blue, the total number of MFs is reported next to the sample name in cyan. Lines in green shades reports the number of MFs in common between two samples. In the line below, in red and blue shades, are reported the percentage of MFs in common on the total number of MFs for each samples. For instance, for the intersection between R10B and R9, 1754 MFs are in common, which represent 77% of the number of MFs in R10B and 70% of the MFs in R9. The higher the two percentages, the higher the similarity of the MFs in the two samples compared. To increase the readability of the table, higher percentages are in red and blue, while low percentages are faded.

At first glance we can see the high similarity of the composition of R8, R9 and R10B, which contain MFs that are also present in the sample 08/10/2021. Samples 02/03/2019 and 15/03/2019 have a quite similar composition and contain more than 1000 MFs detected also in R8, R, R9 and R10B. Nevertheless, this similarity is less visible between the other samples collected at PUY and R10B. Interestingly, even if samples collected at PUY in autumn and summer contain a lower number of MFs, the composition is similar.

In summary, it seems that MFs contained in samples collected during summer and autumn at PUY, with the exception pf 08/10/2021, are different from those observed in samples collected at REU and could justify the different OSC value. Nevertheless, samples collected in winter at PUY are relatively similar to those collected in REU, although the OSC value is significantly different between these samples. Sample 08/10/2021 has such a large variety of MFs that is similar to both samples collected at REU and at PUY.

		R9	R10B	02/03/2019	15/03/2019	02/10/2019	22/10/2019	17/07/2020	03/11/2020	08/10/2021
		2503	2276	3244	2084	1543	120	715	312	7436
R8	3098	2001	1708	1517	1151	701	34	279	108	2274
	% ↑	80%	75%	47%	55%	45%	28%	39%	35%	31%
	% ←	65%	55%	49%	37%	23%	1%	9%	3%	73%
R9	2503		1754	1488	1104	617	32	241	95	1728
	% ↑		77%	46%	53%	40%	27%	34%	30%	23%
	% ←		70%	59%	44%	25%	1%	10%	4%	69%
R10B	2276			1322	1049	584	29	246	96	1477
	% ↑			41%	50%	38%	24%	34%	31%	20%
	% ←			58%	46%	26%	1%	11%	4%	65%
02/03/2019	3244				1722	648	84	295	192	2085
	% ↑				83%	42%	70%	41%	62%	28%
	% ←				53%	20%	3%	9%	6%	64%
15/03/2019	2084					620	85	295	198	1620
	% ↑					40%	71%	41%	63%	22%
	% ←					30%	4%	14%	10%	78%
02/10/2019	1543						107	558	260	1069
	% ↑						89%	78%	83%	14%
	% ←						7%	36%	17%	69%
22/10/2019	120							87	68	98
	% ↑							12%	22%	1%
	% ←							73%	57%	82%
17/07/2020	715								227	531
	% ↑								73%	7%
	% ←								32%	74%
03/11/2020	312									266
	% ↑									4%
	% ←									85%

Lines 362-364: If I am understanding correctly, the general percentage of formulas in each classification is similar between REU and PUY, which seems reasonable, I am still curious about the specific differences

between the molecules in one sample or another in a more comprehensive view. Do the formulas in each classification match each other between the different sites or are they largely different? For example, for the LipidC classification, are the formulas found at REU and PUY 90% common, 70%, 50%, less? I think it could be interesting to see if the detailed composition of these samples is very different or the same, since it may say something about the cloud processing results. The "averages" are very useful, but as you have mentioned, even the same formula doesn't necessarily mean the same molecule, so if a set of molecular formulas are in a particular classification, they may not be similar in any other way, or they could be very similar and highlight that cloud processing brings organic matter to a similar specific result.

We thank the reviewer for this useful comment. Indeed, results are quite surprising for the specific dataset. The comparison of lipids MFs in REU samples highlighted that 48% of the MFs are common to the three samples. Intuitively, we could imagine a similar result for PUY samples: nevertheless, the comparison of lipids MFs shows that only 3% of the total number of lipid MFs (1303) is in common between all the samples. The percentage reaches 4.1% for common MFs in 6 (out of 7) samples, 6.2% for common MFs in 5 samples and 10.3% for common MFs in 4 samples. The comparison between samples collected in PUY and REU is reported in the following table, which shows the number of lipids MFs in each samples (for instance, 4 MFs are present in all ten samples, the total number of lipid MFs (1903) and the % of MFs on the total number. Only 0.2% of the MFs are common to all the samples, showing that lipids from REU are different from lipids from PUY samples.

occurrence in samples	10	9	8	7	6	5	4	3	2
# of MFs	4	28	60	120	356	259	315	379	382
total # of MFs (lipids)	1903								
%	0.2	1.5	3.2	6.3	18.7	13.6	16.6	19.9	20.1

Considering these results, we could argue that cloud processing increases the molecular complexity of cloud water, and we need to look at the final products of cloud processing, such as C1-C3 compounds to find similarity.

A sentence is added in the article.

Lines 386-387: "Although LipidC represents more than half of the MFs in REU and PUY samples, only four MFs of this class are in common between all the samples."

Lines 370: While the FT-ICR is very well suited and effective for this work, the lack of structural information is a shortcoming as noted here, is there any interest in doing LC or fragmentation analysis in the future for these samples or others?

We agree with the reviewer: we need more information on the structure. Nevertheless, it is still difficult to combine LC or fragmentation analysis with FT-ICR MS for complex matrices. A nice work from Divisekara et al., (2023) reports the development of software for this kind of analysis and we would be glad to have the opportunity to test this approach on our samples.

Lines 378-384: You are taking appropriate caution in classifying these molecules as one specific class or another with the database, but I was curious whether if you took a few of the formulas that you have classified as "prenol lipids" for example and just looked for any molecule matching that formula (in other databases or the search engine of your choice) if you could get any other classification?

A quick research of some molecular formula in Chemspider (https://www.chemspider.com/) confirm that we need to interpret our results with extreme caution. For instance, the MF corresponding to myrtenic acid $C_{10}H_{14}O_2$ produced 5609 results in Chemspider and myrtenic is the 80th of the list. Similarly, the MF corresponding to parthenin, $C_{15}H_{18}O_4$, gave 7717 results, with parthenin in 27th position. That's the reason of our caution in this interpretation and why we will be glad to analyze samples in LC-FT-ICR MS.

Lines 426-428: I do not quite understand this sentence. Are the measured concentrations for alpha pinene 0.5, 71.5, and 2 for R8, R9, and R10B, while the beta pinene concentrations were 39.9 and 1.3 for R8, R9, and R10B, or are the detection limits for alpha pinene 39.9 and for beta pinene they are 1.3? I think the sentence could be restructured for clarity.

We agree with the reviewer. The sentence was modified as follows

Lines 451-453: "In particular, alpha pinene concentrations were 0.5, 71.5 and 2.0 nmol L^{-1} and beta pinene concentrations were below detection limit for R8 and 39.9 and 1.3 nmol L^{-1} for R9 and R10B, respectively."

Lines 457: Does this mean that the organosulfate intensity was low in all samples (REU and PUY) with the exception of PUY 8/10/2021, or are you just comparing PUY 08/10/2021 to other PUY samples? Additionally, you explain the higher occurrence of limonene organosulfates at REU by the increased emission of limonene at the site, which makes sense, but does that imply that the organosulfate formation from limonene is a faster process than the oxidation of pinene? My understanding of the reason given for the relative lack of pinene oxidation products is that the emissions were too fresh to have oxidized yet. Is the organosulfate a primary oxidation product like C8H12O5? Or is the explanation that there is more limonene emissions relative to the pinenes?

The organosulfate intensity is higher in the 8/10/2021 sample than in the other samples from the PUY and the REU. The text was modified to clarify the statement. At the REU, limonene emissions are mostly on the coastal region while pinene emissions are prevalent on the slope of the mountain, where samples are collected. That implies that pinene emissions are fresher than limonene emissions. That is the reason why limonene is more oxidized than pinene. We think that the formation of organosulfates in the coastal environment is favored due to the strong emission of NOx from traffic and DMS from the sea, as evidenced in the work from Rocco et al., (2022).

Lines 465: What were the N and S beta caryophalene formulas? Is there any way to know that the formulas are N or S caryophyllene molecules other than matching the formulas? While presence of their emission sources on the coast may explain the N and S beta caryophyllene, why would there be no CHO oxidation products? Are the N and S reactions that much more favorable than the O oxidation? Or is the concentration of N and S so overwhelming that the O oxidation doesn't really occur, relative to N and S?

The hypothesis presented is just a speculation and cannot be supported by the dataset. Thus, it was removed from the text.

Line 524: According to the classification you say that 50% of the molecules observed are reduced, is the explanation that the organic matter in the clouds is fairly fresh and hasn't had a chance to oxidize more completely yet?

This comment is interesting and the hypothesis mentioned could explain the presence of lipids in REU samples. Nevertheless, local sources and an input of fresh organic matter can be excluded at PUY. One

potential explanation is that reduced organic matter, which is likely to be hydrophobic or amphiphilic, such as fatty acids, is located at the interface between water and air and reacts less with photogenerated radicals in the aqueous phase. This can preserve it from the oxidation. When we collect the sample by collision of the droplets on the plates of the collector and coalescence into the bottle, the reduced matter is trapped into the liquid and then, concentrated by SPE and analyzed by FT-ICR MS. This is just a hypothesis that cannot be proved by our results, thus no modifications are reported in the article.

Technical Corrections

Line 25: Somewhat contradictory statements, can consider changing the language a bit to get to the assumed intended meaning.

We agree with the reviewer. The text was modified as follow:

Lines 24-28: "The composition of cloud water dissolved organic matter has been investigated through non-targeted high resolution mass spectrometry only on few samples that were mostly collected in the Northern hemisphere, in USA, Europe and China. There remains, therefore, a lack of measurements for clouds located in the Southern Hemisphere, under tropical conditions and influenced by forest emissions. As a matter of fact, the comparison of the composition of clouds collected in different locations is challenging since the methodology for the analysis and data treatment are not standardized."

Line 179: It may be more consistent and precise to say "same mass" instead of "same peak", since the parenthetical on line 180 says "unique mass".

We agree with the reviewer. The text was modified as follow:

Lines 179-181: The output of the function gives a list of ambiguous (multiple MFs that have been assigned to the same mass) and unambiguous (MFs that have been assigned to a unique mass) MFs.

Lines 323: Should probably change "is" to "are"

We agree with the reviewer. The text was corrected

Lines 520: Instead of "emitted" you should probably say something like "developed" or "produced". Overall the language in this manuscript is very good, but there are few minor things, like this and the comments for lines 426-428 that could be adjusted.

We agree with the reviewer. The text was modified as follow:

Lines 546-548: "We hypothesized that, for autumn samples, strong emissions are rapidly processed at REU, due to the high temperature, and aged air masses are collected at PUY, leading to similar values of average OSC, but produced by different causes."

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