

## Response to referee reports

We thank Maria Kanakidou and the anonymous referee for their constructive comments on our manuscript and their careful referee reports. We respond to their comments below. Referee comments are in blue and our responses in black; modified text is shown in red. All line numbers refer to the revised manuscript version without annotations.

### Maria Kanakidou (Referee)

This is a very interesting paper that provides an innovative view – a new concept – of bacteria in the atmosphere. The authors make the point that a fraction of bacteria in the atmosphere is not inert but can multiply producing secondary bioaerosols and in parallel they can consume water soluble organic mass thus providing an alternative to the chemical degradation path for organic mass. This is a nice and holistic view for the lifecycle of bacteria in the atmosphere. However, there are several gaps of knowledge in this cycle that the authors thoroughly discuss. Based on available literature and a number of assumptions that are clearly stated in the manuscript, the authors make rough calculations to evaluate the two terms involved in the bacteria budget, namely the secondary production of bacteria and the bacteria driven degradation of water soluble organic mass. This later, the biological degradation of organic mass, is compared to the chemical degradation pathway and found of potentially similar importance but with a large range of uncertainty. Despite the significant gaps in knowledge that prohibit accurate estimates, I consider that, overall, the manuscript provides a new concept for the presence and functioning of bacteria in the atmosphere that deserves publication in ACP after some improvements.

We thank the Maria Kanakidou for her positive comments on our manuscript. All comments are addressed in detail below.

Abstract: lines 21-22: ‘ the conditions under which microbial processes cannot be neglected as organic carbon sinks in clouds’ Please provide such information in the abstract.

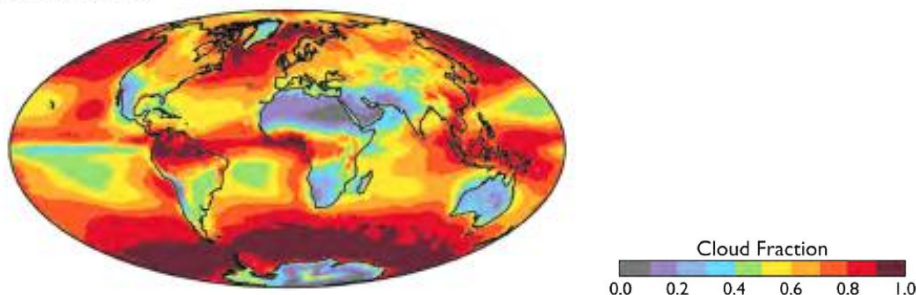
We reworded this sentence as follows (l. 21/22):

*While also this estimate is very approximate, the analysis of the uncertainties and ranges of all parameters suggests **that high concentrations of metabolically active bacteria in clouds might represent an efficient sink for organics.***

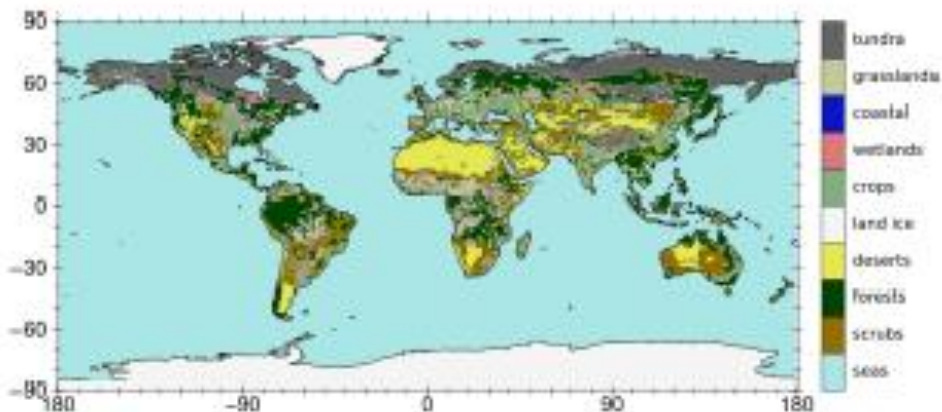
Section 2.4.2: It is unclear which year of MODIS cloud data has been used. It would be nice to show the derived map of cloud volume and the ecosystem map that are later used to derive the numbers in Table 2 (in page 8 that is erroneously numbered as Table 1). What grid size is used for these calculations? How many grid points are used to derive the  $F_{\text{cloud}}$  over each ecosystem type? This is an important Table for the budget estimates that are further presented in the manuscript; therefore, it has to be well documented.

We agree that our description of the MODIS data and cloud cover was too short (cf also our response to the comments by the other referee). We used data for spring (March – May) of MODIS Terra data, averaged over twelve years (2000 – 2011). We visually overlaid the map (Figure R-1) to the map by Burrows et al. (2009b), Figure R-2)

(b) March-May



**Figure R-1: Seasonal mean daytime cloud fraction from Terra (2000-2011) for March – May, this figure is Figure 2b in King et al., (2013)**



**Figure R- 2: Lumped ecosystem classes, based on the Olson World Ecosystems (Olson et al., 1992); Figure 1 in Burrows et al., 2009b.**

This way, we estimated the cloud cover for the major ecosystems as listed by Burrows et al. (2009), based on the categories defined by Olson et al. (1992). We agree with the referee that a more detailed view could be used to characterize small scale features. However, given the conceptual nature of our study that builds upon the categories as used in the previous study by Burrows et al. (2009) for primary bacteria emissions, we think that our approach is sufficient to give (i) a reasonable estimate of cloudiness above the various ecosystems and (ii) sufficient detail of its concept to be refined in future studies.

Adding also a column with cloud fraction over each ecosystem as suggested by the other reviewer will be a significant improvement.

We extended Table 2 and added the cloud cover  $F_{clc}$ .

**Table 2:** Surface coverage of ecosystems on Earth surface (Burrows et al., 2009b), *approximate cloud coverage  $F_{clc}$  above the ecosystems, estimated based on maps of annual cloud cover data obtained by MODIS, and estimated time fraction bacteria spend in clouds ( $F_{cloud}$ )*

	<b>% of Earth surface</b> a)	<b><math>F_{clc}</math></b>	<b><math>F_{cloud}</math></b>
All	100	0.6	0.15
Tundra	3.3	0.4	0.1
Grassland	2.2	0.7	0.2
Coastal	0.2	0.4	0.1
Wetlands	0.6	0.5	0.15
Crops	3.0	0.7	0.3
Land ice	3.1	0.4	0.1
Deserts	3.7	0.2	0.05
Forests	7.0	0.9	0.25
Shrubs	5.8	0.3	0.1
Seas	71.0	0.7	0.2

a) Data from Burrows et al. (2009b) and Olson (1992)

It is also unclear

1) how the value of 0.15 is derived for  $F_{Cloud}$ ,

We modified the previous text as follows (l. 181ff):

In general, cloud contact times, i.e., the time air spends in a cloud, are dependent on cloud depth and vertical velocity (Feingold et al., 2013). This small-scale information is not consistently available for the large regions as covered by the ecosystems listed in **Table 1**. ~~Instead, the fraction of cloud contact time  $F_{cloud}$  is here equaled to the atmospheric volume fraction that is filled with clouds (Lelieveld and Crutzen, 1990).~~

~~Using global cloud coverage data based on satellite images derived from MODIS data (King et al., 2013) and an average cloud thickness of 1.5 km within the lowest 6 km of the atmosphere (Pruppacher and Jaenicke, 1995; Wang et al., 2000), we apply a factor of 0.25 (1.5 km / 6 km) to obtain an average cloud volume, and thus  $F_{cloud}$ , above each ecosystem. In order to give an estimate of the cloud processing time over the various large ecosystems as identified by Olson et al. (1992), we use the approximate cloud fractions during spring averaged for 2000 - 2011 from MODIS Terra (e.g., Figure 2b in (King et al., 2013)). While this representation gives only some snapshot of cloudiness that varies over smaller spatial and temporal scales. However, given the conceptual nature of our study that builds upon the categories as used in the previous study by Burrows et al. (2009) for primary bacteria emissions, our approach seems sufficient to give (i) a reasonable estimate of cloudiness above the~~

various ecosystems and (ii) enough detail of its concept to be refined in future studies.

Globally, a range of cloud thicknesses of 1.4 – 1.9 km has been derived (Table 1 in (Wang et al., 2000)) from which we use  $h = 1.5$  km as a single value for the average cloud thickness. Assuming further that globally > 90% of all liquid clouds reside in the lowest 6 km of the atmosphere ( $\Delta z = 6$  km) (Pruppacher and Jaenicke, 1995), we can convert the cloud coverage as obtained from satellite data into cloud volume fractions using Eq-x:

$$F_{cloud} = F_{clc} \cdot \frac{\text{cloud thickness [km]}}{\Delta z \text{ [km]}} = F_{clc} \cdot \frac{1.5 \text{ km}}{6 \text{ km}} \quad (3)$$

Comparison of previous estimates of global cloud coverage of 60% (Pruppacher and Jaenicke, 1995) and the volume fraction of liquid clouds within the atmosphere of 15% (Lelieveld and Crutzen, 1990) generally supports this relationship. The resulting  $F_{cloud}$  values are summarized in **Table 2** together with the percentage area fraction of each ecosystem of the Earth surface, taken from Burrows et al. (2009b) and originally obtained from Olson (1992), and the cloud coverage data.

2) whether the category ‘Seas’ in Table 2 contains also the sea-ice.

No, the category ‘land-ice’ does not include sea-ice. The categories by Olson et al., do not include sea-ice as a separate category. There is no data available for bacteria cell concentrations above sea ice; however, we assume that cell concentrations and microbial activity are low there due to the lack of primary sources and because of low temperatures, respectively. We added this information to the text (l. 224ff):

According to the definition of the categories as suggested by Olson et al. (1992), the category ‘land ice’ does not include sea ice. It can be expected that above sea ice the sources and metabolic activity of bacteria are also very low (Martin et al., 2009) and thus can be likely neglected on a global scale.

Line 215: explain to what you refer when writing ‘other formation rates

We reworded the text and replaced ‘other formation rates’ by ‘formation rates in the individual ecosystems’. (l. 221)

‘Caption of figure 3. The reference to Burrows et al is incomplete.

We completed the reference.

Line 273: ‘sensitivity’ I think ‘uncertainty’ is more appropriate here.

We agree. We replaced ‘sensitivity’ by ‘uncertainty’ (l. 285).

Table 3: maximum range for  $C_{cell}$  (1E9) can you comment who and where has measured this ?

**A:** As indicated as a comment in the Table, this value of  $C_{cell}$  originates from the work by Paez-Rubio et al. (2005) and it is considered the highest expectable value for the concentration of airborne bacteria. This was observed above the surface of an agricultural field irrigated with nondisinfected effluent from a wastewater

storage lagoon in Mexico. Aerosol sampling was performed with biosamplers (impingers) and enumeration of total bacteria was done by epifluorescence microscopy observation. We added information about this particular location in Table 3 and in more detail in l. 253:

$C_{cell,max}$  was measured ~~near the ground~~ above a wastewater storage lagoon and can be considered the highest expectable value of ambient bacteria (Paez-Rubio et al., 2005)

Table 3: comment for mcell first line: ‘assuming that they’ to avoid confusion replace ‘they’ by ‘cells’.

As suggested, we clarified the sentence and replaced ‘they’ by ‘cells’.

Lines 283-285: discussion about fungal spores: I do not see why this is discussed here. Please remove or rephrase sentence to better fit in the discussion

**A:** As the section starts out with discussing total primary biological particles, we think that mentioning fungal spores here is relevant as they contribute a major fraction to total PBA. We agree with the reviewer that the wording was not clear and thus we modified the text as follows (l. 294ff):

Bacteria usually comprise only a small mass fraction of total PBA; a major fraction is composed of fungal spores. Thus, their emissions are generally estimated to be larger in mass than those of bacteria. ~~For example, fungal spores are a major fraction of PBA. Their~~ global emissions (25 Tg yr<sup>-1</sup>) was suggested to contribute 23% to total primary organic aerosol (Heald and Spracklen, 2009). An estimate of fungal spore emissions based on tracer compounds resulted in predicted 50 Tg yr<sup>-1</sup> (Elbert et al., 2007)

Equation 6:  $F_c$  needs to be defined earlier, now it is defined in line 339.

We agree that the definition of  $F_c$  should have been included right after Equation (7). We moved the text to l. 348:

Equation 7 includes an additional factor  $F_c$  ~~in Eq (5)~~ that accounts for the microbial selectivity towards only some organics by each bacteria type (e.g. Šantl-Temkiv et al., 2013; Bianco et al., 2019).

Also check units in this equation.

We thank the referee for noticing the mistake in the equation. We corrected it as follows:

$$R_{WSOC,Bact} \left[ \frac{g_c}{L(aq)_s} \right] = -F_{CO_2} k_{Bact} \left[ \frac{L(aq)}{cell \cdot s} \right] \cdot C_{Cell,aq} \left[ \frac{cell}{L(aq)} \right] \cdot F_c \cdot C_{WSOC} \left[ \frac{g_c}{L(aq)} \right] \cdot F_{cloud} \quad (6)$$

Whereas the cell concentration in cloud water can be replaced by

$$C_{Cell,aq} \left[ \frac{cell}{L(aq)} \right] = C_{cell,g} \left[ \frac{cell}{m^3(g)} \right] / LWC \left[ \frac{m^3(g)}{L(aq)} \right] \quad (7)$$

with  $C_{Cell,aq}$  and  $C_{WSOC}$  being the concentrations of bacteria and water-soluble organic carbon in cloud water, respectively, and  $C_{cell,g}$  ambient cell concentrations in the gas phase (e.g. Table 1).

Line 340-342: provide an uncertainty range for  $F_c$  instead of one value. How this uncertainty is affecting the here presented estimates of WSOC loss by bacteria?

To our knowledge,  $F_c$  or any similar parameter has not been determined experimentally on ambient cloud water or aerosol samples.  $F_c$  is the fraction of organic material that can be metabolized by bacteria in clouds (i.e. the potential of organic compounds to be biodegraded, which depend on their bioavailability and chemical formulae and structure). In general, only a small fraction of all cloud water organics (~15%) can be speciated on a molecular basis (Herckes et al., 2013). We added to the manuscript the text below (l. 354ff) as the justification of  $F_c = 0.5$ . As we discuss a range of  $F_c$  (0.2 – 0.8) in Table 4, we refrained from adding a range here in the text.

Given the complexity of the organic matter in the atmosphere, the numerous organic molecules existing in cloud water and their variable susceptibility to biodegradation, this fraction is hard to specify with precision. Bianco et al. (2019) observed experimentally by FT-ICR-MS during laboratory incubation of cloud water that out of the 2178 compounds detected, 1094 were actually utilized by bacteria (~ 50%). Assuming that all these compounds were equally abundant, one could conclude that 50% of all cloud water organics were prone to be microbiologically consumed (i.e.  $F_c = 0.5$ ). More quantitative support of this assumption could be given based on the fact that preferably small oxygenated organics are taken up by bacteria. Compilations of speciated cloud water organics have shown that small acids, such as formic and acetic acid, comprise a large fraction (up to ~30%) of the characterized fraction of cloud water organics (e.g. Figure 6, (Herckes et al., 2013). Lab experiments have shown that these acids are degraded by bacteria (Amato et al., 2007; Vaitilingom et al., 2013).

Line 364: 'slightly higher contribution of chemical reactions to WSOC loss'. Figure 4 shows that for  $Y_{voc}$  equal 0.5 the chemical loss can be double the bacteria loss. This is not 'slightly higher'. Please rephrase.

We rephrased the text as follows (l. 385ff):

The results in **Figure 4** ~~show~~ suggest that the microbial rates may be smaller than the chemical ones under most conditions. Overall, the values shown in Figure 4 only differ by a factor of ~2.5 which might imply that there are conditions under which chemical and biological processes in the aqueous phase compete.

~~predicted rates  $R_{chem,WSOC}$  (8 – 20 Tg yr<sup>-1</sup>) and  $R_{bact,WSOC}$  (8 – 11 Tg yr<sup>-1</sup>) are for most parameter sets on a similar order of magnitude with possibly a slightly higher contribution of chemical reactions to WSOC loss.~~

Line 372: Loss rate of 50 Tg/yr is stated here while in Figure 4, a maximum of about 30 Tg/yr is calculated. Make consistent.

**A:** Thank you for noticing this typo mistake. We modified to ~30 Tg/yr, consistently with Figure 4 (l. 396)

Line 377: the authors claim that the WSOC losses are smaller than the predicted production rates of SOA<sub>aq</sub>. However, when accounting for the range of these rates, there is no significant difference. Furthermore, this result will depend on the assumed  $F_c$ , so please rephrase.

We agree with the reviewer that our statements were not accurate. Part of this is due to the fact that aqSOA and WSOC are not necessarily comparable. There is clearly an overlap between these two groups of organics; however, they do not denote the same mass as WSOC might include volatile compounds that do not contribute to aqSOA after drop evaporation and not all aqSOA might be water soluble.

We added (l. 401ff):

For the parameters chosen in our estimate, they are also smaller than the predicted production rates of aqSOA in clouds of 13.1 - 46.8 Tg year<sup>-1</sup> (Lin et al., 2014) or 20 - 30 Tg yr<sup>-1</sup> (Liu et al., 2012). This estimate represents organic carbon sources and sinks in general. It should be noted that the organics included in WSOC and aqSOA, respectively, might not be identical.

Line 433: I think 0.7 Tg/yr should be 3.7 Tg/yr

We agree with the referee that the reference to the average value of 0.7 Tg/year is confusing. As we had stated before, we present here the value of 3.7 Tg/year as the 'best estimate' and replaced the number accordingly (l. 459)

Line 458: in our study provides.

Thanks for noticing this. We removed 'is' in the sentence.

In addition, please provide references for

1) Figure 1 in its caption

**A:** The schematic in Figure 1 is conceptual of the fact that bacteria either grow in size, divide, stay dormant or die. There is no specific and unique reference for this, so we included the following references: (Si et al., 2017)(Norris, 2015) for bacteria cells growing in size and dividing, (Kaprelyants et al., 1993; Price and Sowers, 2004) for cell dormancy, and (Engelberg-Kulka et al., 2006) for cell death.

We added in l. 67ff:

whereas bacteria dormancy and death do not lead to any change in cell mass (Kaprelyants and Kell, 1993; Price and Sowers, 2004)

2) the value of 7 to 14% in Line 159.

The values of 7-14% for BGE were taken from the reference cited in the sentence before (Eiler et al., 2003). In order to make it clearer, we connected the two sentences by (l. 158)

BGE for planktonic bacteria range from < 0.4% to 80% with the highest values for eutrophic conditions (Eiler et al., 2003). In turn, in the same study, it was shown that, when substrate availability is limited, values from ~7% to ~14% are generally observed.

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### Reviewer #3

This is a very interesting and important study that identifies and tackles a major gap in the aerosol cloud interactions, that is already lacking primary biological particles to a large extent and comes up with some rough estimates of the secondary biological particles. I find the paper suitable to be published in ACP, given that some issues raised below are answered.

We thank the referee for his/her positive evaluation of our manuscript. All comments are addressed in detail below.

1) How about sea-ice? Is it considered together with land-ice or not considered at all?

Sea ice is not considered in the major categories as defined by Olson et al. (1992) and as used in the global model study on primary bacteria emissions by Burrows et al. (2009). We added (l. 224ff):

According to the definition of the categories as suggested by Olson et al. (1992), the category 'land ice' does not include sea ice. It can be expected that above sea ice the sources and metabolic activity of bacteria are also very low (Martin et al., 2009) and thus can be likely neglected on a global scale.

2) Similarly, urban sources? Why are not they represented as they can be a large source of bacteria due to human existence?

We focused on the large-scale ecosystems as suggested in the framework by Burrows et al. that, in turn, was based on the ecosystem categories defined by Olson. We agree with the referee that urban areas might be a particular source of bacteria and added the following text (l. 251ff):

However, spatial deviations might be present in particular locations, such as cell concentrations of  $\sim 7 \cdot 10^5 - 4 \cdot 10^6 \text{ cells m}^{-3}$   ~~$\sim 7 \cdot 10^6 \text{ cells m}^{-3}$~~   ~~$10^6 \text{ m}^{-3}$~~  that were found in highly polluted areas during haze periods in Chinaas (Li et al., 2018; Xie et al., 2018), and even  $10^9 \text{ m}^{-3}$  above a wastewater storage lagoon which can be considered the highest expectable value of ambient bacteria near the ground (Paez-Rubio et al., 2005). Using the framework presented in the present study, SBA formation in such rather spatially limited areas can be estimated if growth rates of the individual bacteria types were available.

Generally, we'd like to note that this study aims at providing a first estimate of the contribution of bacteria to secondary biological aerosols (SBA), and providing a frame for future investigations in case where mode specific cases need to be studied. For now, this only includes the major ecosystems; given the general lack of data about bacteria abundance and activity in the atmosphere, and the large uncertainties associated with bacteria emissions (see Tables 3 and 4) from surfaces, we indeed intentionally simplified the global system. However, we completely agree that including more detailed information would be highly interesting and

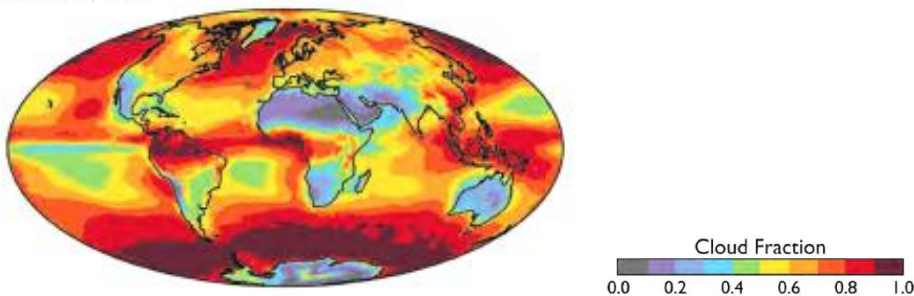


needed, so we hope that our study will give in the model in the future and explore different scenarios of bacteria emission, cloud cover, temperature, etc.

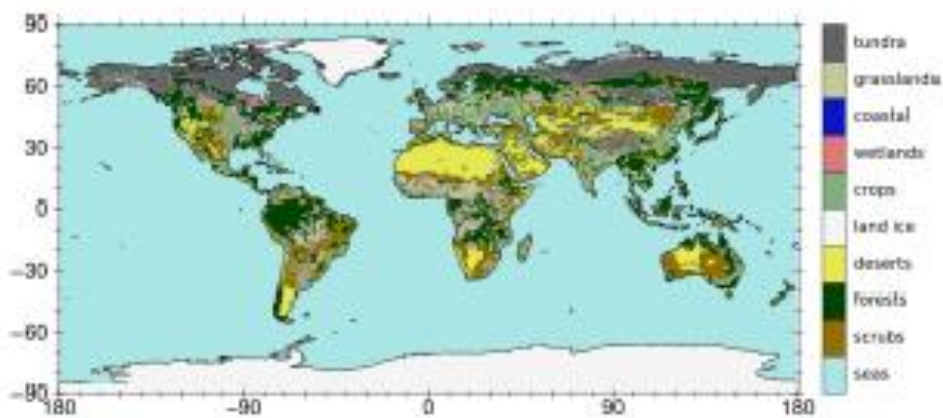
3) Is it possible to provide with a formula that calculates  $F_{\text{cloud}}$  based on ecosystem, corresponding cloud fraction from MODIS and the conversion factor in order to be able to reproduce the values in Table 2? Table 2 can be updated to include the cloud fraction over each ecosystem.

We agree that our description of the MODIS data and cloud cover was too short (cf also our response to the comments by the other referee). We used data for spring (March – May) of MODIS Terra data, averaged over twelve years (2000 – 2011). We visually overlaid the map (Figure R-1) to the map by Burrows et al. (2009b), Figure R-2)

(b) March-May



**Figure R-3: Seasonal mean daytime cloud fraction from Terra (2000-2011) for March – May, this figure is Figure 2b in King et al., (2013)**



**Figure R- 4: Lumped ecosystem classes, based on the Olson World Ecosystems (Olson et al., 1992); Figure 1 in Burrows et al., 2009b.**

This way, we estimated the cloud cover for the major ecosystems as listed by Burrows et al. (2009), based on the categories defined by Olson et al. (1992). We agree with the referee that a more detailed view could be used to characterize small scale features. However, given the conceptual nature of our study that builds

upon the categories as used in the previous study by Burrows et al. (2009) for primary bacteria emissions, we think that our approach is sufficient to give (i) a reasonable estimate of cloudiness above the various ecosystems and (ii) sufficient detail of its concept to be refined in future studies. We modified the text as follows (l. 181ff):

In general, cloud contact times, i.e., the time air spends in a cloud, are dependent on cloud depth and vertical velocity (Feingold et al., 2013). This small-scale information is not consistently available for the large regions as covered by the ecosystems listed in **Table 1**. ~~Instead, the fraction of cloud contact time  $F_{cloud}$  is here equaled to the atmospheric volume fraction that is filled with clouds (Lelieveld and Crutzen, 1990).~~

~~Using global cloud coverage data based on satellite images derived from MODIS data (King et al., 2013) and an average cloud thickness of 1.5 km within the lowest 6 km of the atmosphere (Pruppacher and Jaenicke, 1995; Wang et al., 2000), we apply a factor of 0.25 (1.5 km / 6 km) to obtain an average cloud volume, and thus  $F_{cloud}$ , above each ecosystem. In order to give an estimate of the cloud processing time over the various large ecosystems as identified by Olson et al. (1992), we use the approximate cloud fractions during spring averaged for 2000 - 2011 from MODIS Terra (e.g., Figure 2b in (King et al., 2013)). While this representation gives only some snapshot of cloudiness that varies over smaller spatial and temporal scales. However, given the conceptual nature of our study that builds upon the categories as used in the previous study by Burrows et al. (2009) for primary bacteria emissions, our approach seems sufficient to give (i) a reasonable estimate of cloudiness above the various ecosystems and (ii) enough detail of its concept to be refined in future studies.~~

Globally, a range of cloud thicknesses of 1.4 - 1.9 km has been derived (Table 1 in (Wang et al., 2000)) from which we use  $h = 1.5$  km as a single value for the average cloud thickness. Assuming further that globally > 90% of all liquid clouds reside in the lowest 6 km of the atmosphere ( $\Delta z = 6$  km) (Pruppacher and Jaenicke, 1995), we can convert the cloud coverage as obtained from satellite data into cloud volume fractions using Eq-x:

$$F_{cloud} = F_{clc} \cdot \frac{\text{cloud thickness [km]}}{\Delta z [km]} = F_{clc} \cdot \frac{1.5 \text{ km}}{6 \text{ km}}$$

Comparison of previous estimates of global cloud coverage of 60% (Pruppacher and Jaenicke, 1995) and the volume fraction of liquid clouds within the atmosphere of 15% (Lelieveld and Crutzen, 1990) generally supports this relationship. The resulting  $F_{cloud}$  values are summarized in **Table 2** together with the percentage area fraction of each ecosystem of the Earth surface, taken from Burrows et al. (2009b) and originally obtained from Olson (1992), and the cloud coverage data.

4) Table 1 caption in section 2.4.2 should be corrected to Table 2.

We thank the referee for noticing this. We corrected the typo.

5) Is it possible to distinguish the different forest types or regions? It would be interesting to see these numbers above the amazons and boreal forests for example. Therefore, it would be interesting to show that global spatial distribution of this SBA source.

We fully agree that a finer spatial resolution of the individual ecosystems or subsystems would be interesting. However, the data sets of measured bacteria cells above different regions are rather limited. In the boreal forests near Hyytiälä, Finland, bacteria cell concentrations of  $6323 \pm 13748 \text{ m}^{-3}$  were found (Helin et al., 2017), which is very close to the average concentration of  $10000 \text{ cell m}^{-3}$  shown in Table 1. Using an average cloud cover of 0.65 for boreal forests as derived by (Spracklen et al., 2008) translates into a cloud volume fraction of  $\sim 0.15$  using the equation employed here. Thus, in a first approximation, it can be expected that the SBA source from boreal forests [ $\text{ng m}^{-3}$ ] may be similar to the average one suggested here. Above the pristine forests of the Amazon, a rich biodiversity of prokaryotes has been found (Souza et al., 2019). However, in that study, the cell concentrations were not quantified. Given the large cloud cover of  $> 75\%$  during the wet season in this area (Marquardt Collow et al., 2016), it can be expected that SBA formation might be significant there. Similarity, in the tropics, the great diversity of bacteria was characterized qualitatively (Gusareva et al., 2019); however, also in this latter study, no quantitative data on ambient cell concentration was given. In a study by (Huffman et al., 2012) a small mode of biological particles between  $0.5 - 1 \text{ microm}$  was identified during the AMAZE-08 campaign in the Amazon rain forest that likely contained bacteria. However, that study did not quantify them further. Preliminary data suggest concentrations of bacteria cells of  $10^4 - 10^5 \text{ m}^{-3}$  (personal communication, C. Pöhlker).

In order to highlight the lack of quantitative measurements of cell concentrations in the aforementioned regions and in general, we added the following text (l. 466ff):

The ecosystem categories in Table 2 represent fairly large regions. It might be expected that SBA formation rates are different on smaller spatial and/or temporal scales. For example, it has been shown that human activities in cities lead to high bacteria concentrations; also forests have been identified as significant sources of biogenic aerosol. However, detailed data on bacteria are sparse in such regions. While several recent studies have characterized the diversity of microorganisms in forested regions (rainforest, tropics) (Gusareva et al., 2019; Souza et al., 2019), these studies did not report cell concentrations which highlights the urgent need of additional measurements.

#### 6) Line 236: 1% of the secondary aerosols.

We assume that the referee referred to line 296 ('Thus, SBA production can be estimated to be on the order of  $\sim 1\%$  of the total aerosol sources'). We reworded as follows (l. 308):

Thus, SBA production can be estimated to be on the order of  $\sim 1\%$  of the secondary aerosol sources.

#### 7) Line 343: Where does the $F_c=0.5$ value come from, any reference or argument

To our knowledge,  $F_c$  or any similar parameter has not been determined experimentally on ambient cloud water or aerosol samples.  $F_c$  is the fraction of organic material that can be metabolized by bacteria in clouds (i.e. the potential of organic compounds to be biodegraded, which depend on their bioavailability and chemical formulae and structure). In general, only a small fraction of all cloud water organics (~15%) can be speciated on a molecular basis (Herckes et al., 2013). We added to the manuscript the text below as the justification of  $F_c = 0.5$ . As we discuss a range of  $F_c$  (0.2 – 0.8) in Table 4, we refrained from adding a range here in the text. (l. 354ff):

Given the complexity of the organic matter in the atmosphere, the numerous organic molecules existing in cloud water and their variable susceptibility to biodegradation, this fraction is hard to specify with precision. Bianco et al. (2019) observed experimentally by FT-ICR-MS during laboratory incubation of cloud water that out of the 2178 compounds detected, 1094 were actually utilized by bacteria (~ 50%). Assuming that all these compounds were equally abundant, one could conclude that 50% of all cloud water organics were prone to be microbiologically consumed (i.e.  $F_c = 0.5$ ). More quantitative support of this assumption could be given based on the fact that preferably small oxygenated organics are taken up by bacteria. Compilations of speciated cloud water organics have shown that small acids, such as formic and acetic acid, comprise a large fraction (up to ~30%) of the characterized fraction of cloud water organics (e.g. Figure 6, (Herckes et al., 2013). Lab experiments have shown that these acids are degraded by bacteria (Amato et al., 2007; Vaithilingom et al., 2013).

## References

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# The global impact of bacterial processes on carbon mass

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**Abstract.** Many recent studies have identified biological material as a major fraction of ambient aerosol loading. A  
10 small fraction of these bioaerosols consist of bacteria that have attracted a lot of attention due to their role in cloud  
formation and adverse health effects. Current atmospheric models consider bacteria as inert quantities and neglect cell  
growth and multiplication. We provide here a framework to estimate the production of secondary biological aerosol  
(SBA) mass in clouds by microbial cell growth and multiplication. The best estimate of SBA formation rates of 3.7  
15 Tg yr<sup>-1</sup> ~~are-is~~ comparable to previous model estimates of the primary emission of bacteria into the atmosphere, and  
thus might represent a previously unrecognized source of biological aerosol material. We discuss in detail the large  
uncertainties associated with our estimates based on the rather sparse available data on bacteria abundance, growth  
conditions and properties. Additionally, the loss of water-soluble organic carbon (WSOC) due to microbial processes  
in cloud droplets has been suggested to compete under some conditions with WSOC loss by chemical (OH) reactions.  
Our estimates suggest that microbial and chemical processes might lead to a global loss of WSOC of 8 - 11 Tg yr<sup>-1</sup>  
20 and 8 - 20 Tg yr<sup>-1</sup>, respectively. While also this estimate is very approximate, the analysis of the uncertainties and  
ranges of all parameters ~~gives hints about the conditions under which microbial processes cannot be neglected as  
organic carbon sinks in clouds~~ suggests that high concentrations of metabolically active bacteria in clouds might  
~~represent an efficient sink for organics~~. Our estimates also highlight the urgent needs for more data concerning  
microbial concentrations, fluxes and activity in the atmosphere to evaluate the role of bacterial processes as net aerosol  
25 sink or source on various spatial and temporal scales.

## 1. Introduction

The characterization and quantification of outdoor bioaerosols is an active field of current atmospheric research since  
bioaerosols have been suggested to contribute to adverse health effects and cloud formation as ice-nucleating particles  
(Després et al., 2012). Biological material includes debris, pollen, bacteria, fungal spores, and viruses and is usually  
30 considered as being directly emitted to the atmosphere (primary biological aerosol, PBA (Jaenicke, 2005)). The total  
number and mass concentrations of PBA particles vary widely in space and time: Posfai et al. (1998) found 1% of  
particles with biological material above the Southern Ocean whereas Artaxo et al. (1990) identified more than 90% of  
all particles to contain biological material during the wet season in the Amazon. In an urban/remote region in Germany,



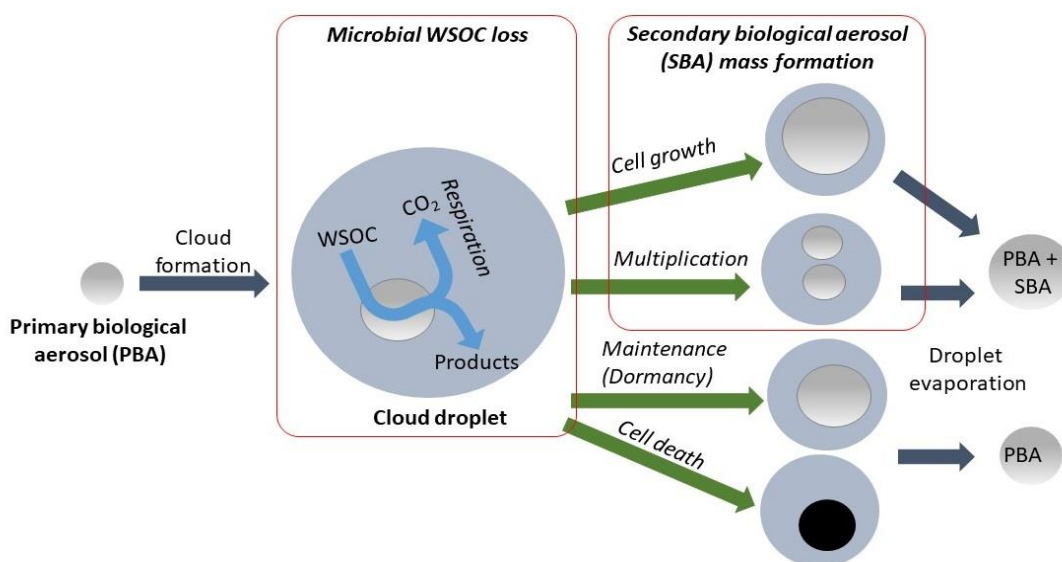
24% of all particles were found to include a biological fraction (Matthias-Maser and Jaenicke, 2000). Similar concentrations were observed at a remote high-altitude site with 16 - 64% of the mass of particles with diameters of less than 10  $\mu\text{m}$  being composed of biological mass (Wiedinmyer et al., 2009) whereas the PBA number fraction was much smaller (0.3 - 18%) in Rome, Italy (Perrino and Marcovecchio, 2016). Bacteria only comprise a small fraction of the total biological aerosol mass but they alone can contribute up to about ~20% of the total number of particles with diameters greater than 0.5  $\mu\text{m}$  (Bowers et al., 2012).

Near the ground, typical concentrations of total airborne bacteria range from ~100 to  $10^6$  cells  $\text{m}^{-3}$ , depending on the emission source (Burrows et al., 2009b), and on temporal, meteorological, and other environmental conditions influencing its propensity to emit particles to the air (Carotenuto et al., 2017; Huffman et al., 2013; Lighthart, 1997; Lighthart and Shaffer, 1995). Atmospheric mixing aloft tends to homogenize the number and diversity of the various bacteria types as the distance from sources increases. In the free troposphere, concentrations of ~10,000 cells  $\text{m}^{-3}$  are reported, including in clouds (DeLeon-Rodriguez et al., 2013; Väitilingom et al., 2013). Some extent of selection toward certain species of bacteria probably occurs during aerosolization and atmospheric transport (Joly et al., 2015; Michaud et al., 2018). However, such selection has not been clearly proven yet as the bacterial assemblages found at high altitude often resemble those observed near the ground (Amato et al., 2017; DeLeon-Rodriguez et al., 2013; Smith et al., 2018).

The atmosphere is a harsh environment for living microorganisms: low temperatures at high altitude, UV radiation (Madronich et al., 2018) and high free radical concentrations (Haddrell and Thomas, 2017; Marinoni et al., 2011) are thought to greatly challenge living organisms (Amato et al., 2019; Joly et al., 2015; Smith et al., 2011). Additionally, the rapidly changing conditions in clouds, like condensation/evaporation and freeze/thaw cycles, can cause strong physiological shocks and physical damages to cells, which can eventually be lethal. The viability of airborne microorganisms is thus very variable in space and time depending on environmental conditions (Fahlgren et al., 2010; Hu et al., 2017; Lighthart and Shaffer, 1995; Monteil et al., 2014), but yet the fact that a fraction of bacteria cells are viable was shown in many experiments of microbiological cultures from ambient aerosol samples (Amato et al., 2007b; Bovallius et al., 1978; Lighthart, 1997; Newman, 1948). This was specified and quantified more recently by direct observations and measurements of biological activity imprints (Amato et al., 2007a, 2017; Sattler et al., 2001; Wirgot et al., 2017). The multiplication of airborne bacteria was observed from aerosols generated from bacteria cultures (Dimmick et al., 1979), as well as in natural polluted fog (Fuzzi et al., 1997). Thus, the estimated **PBA** emissions might be biased high as SBA formation provides an additional source of bacteria mass and, thus, observed bacteria concentrations represent the sum of emission fluxes that are smaller than assumed and the secondary production in the atmosphere.

Efficient bacteria cell growth and multiplication are largely constrained by the presence of liquid water (Davey, 1989; Haddrell and Thomas, 2017). One can thus assume that microbial processes in the atmosphere are limited to the time microorganisms spend in clouds (**Figure 1**). Cell growth and multiplication lead to an increase of the initial cell mass and, thus, to more biological material (Kaprelyants and Kell, 1993; Norris, 2015; Si et al., 2017) whereas bacteria dormancy and death do not lead to any change in cell mass (Engelberg-Kulka et al., 2006; Kaprelyants and Kell, 1993;

70 [Price and Sowers, 2004](#)—). We introduce the term ‘secondary biological aerosol’ (SBA) mass here in order to distinguish this aerosol source from directly emitted PBA. Heterotrophic bacterial processes require the uptake of organic substrates by the cells, which are subsequently converted by metabolic processes into new organic products, biochemical energy and CO<sub>2</sub> (‘respiration’, **Figure 1**). These substrates include organics (e.g., carboxylic acids, sugars); other elements (e.g., nitrogen, phosphorous, potassium, metals) are also needed and exist in bioavailable forms in cloud water. The biotransformation of formate, acetate, succinate, lactate, oxalate, and formaldehyde (Ariya et al., 2002; Vaithilingom et al., 2010), phenol (Lallement et al., 2018) and methane (Šantl-Temkiv et al., 2013) by bacteria and fungi was studied in aqueous solution mimicking the typical chemical composition of cloud water, and it was suggested that under specific conditions, microbial processes might be competitive to chemical radical processes as sinks for these compounds (Delort et al., 2010; Vaithilingom et al., 2011, 2013). The efficiency of such metabolic processes strongly depends on the bacteria types, substrates and their availability within the cloud droplets. In the present study, we perform an estimate of the global importance of SBA formation and microbial WSOC loss. All parameters and their uncertainties are discussed based on the sparse data sets currently available.



**Figure 1:** Bacterial processes in the atmosphere leading to SBA formation and loss of water-soluble organic carbon (WSOC) in clouds.

## 2. Data and assumptions on bacterial processes in clouds

### 85 2.1 Atmospheric concentrations of bacteria cells

Burrows et al. (2009a, 2009b) have summarized data on number concentrations and emission fluxes of bacteria above various ecosystems on the Earth surface. Their compilation also ~~included~~ includes the estimates of cell concentrations near the surface in a range of 10,000 m<sup>-3</sup> (seas) up to 650,000 m<sup>-3</sup> (urban). Bacteria populations aloft represent a mixture of bacteria that were emitted from different ecosystems and subsequently mixed (Burrows et al., 2009b).

90 Despite these mixing processes, there are bacteria types that can be considered characteristic for each ecosystem (Wéry

et al., 2017). **Table 1** lists cell concentrations as published by Burrows et al. (2009b) complemented by some more recent measurements. We extend this overview by data on bacteria types, suggested as predominant or characteristic for each ecosystem. In several cases, more than one predominant bacteria type is listed as specific geographical, meteorological and other environmental conditions might lead to differences in the diversity of bacteria populations for the same category of ecosystem. We also provide global average data (Category ‘All’) and define one of the most abundant bacteria type (Alpha-Proteobacteria) alive in the atmosphere (Amato et al., 2019; Klein et al., 2016) as a representative type. ~~The atmospheric lifetime of bacteria cells is limited to several minutes (Otero Fernandez et al., 2019) to hours (Amato et al., 2015).~~ Several studies report total concentrations of bacteria cells in the atmosphere whereas others present only the concentration of viable cells. The complexity of distinguishing viable, cultivable and dead bacteria cells in the atmosphere has been discussed in several studies (Burrows et al., 2009a; Otero Fernandez et al., 2019).

**Table 1:** Summary of ambient cell concentrations  $C_{cell}$  and generation rates  $R_{cell}$  for predominant bacteria types in all ecosystems

	$C_{Cell} [m^{-3}]^a$	<b>Representative strain affiliation</b>	<b>Generation rate <math>R_{Cell} [h^{-1}]</math></b>
All	10,000	Alpha-Proteobacterium <i>Sphingomonas</i> sp. (average of 32b-11, 32b-49, 32b-57, 35b-32, 35b-38) <sup>e</sup>	0.06 (5°C) <sup>e</sup> 0.2 (17°C) <sup>e</sup> 0.35 (27°C) <sup>e</sup> 0.45 (37°C) <sup>e</sup>
Tundra	12,000	<i>Pseudomonas</i> spp. ( <i>P. graminis</i> ) <sup>e, f, g</sup>	0.12 (5°C) <sup>e</sup> 0.21 (17°C) <sup>e</sup> 0.82 (27°C) <sup>e</sup> 0.27 (37°C) <sup>e</sup>
Grassland	110,000	<i>Psychrobacter</i> sp. <sup>g</sup> <i>Rhodococcus</i> sp. <sup>g</sup> <i>Pseudomonas syringae</i> <sup>e, h</sup>	0.0007 (-10°C) <sup>g</sup> 0.0001 (-10°C) <sup>g</sup> 0.1 (5°C) <sup>e</sup> 0.25 (15°C) <sup>e</sup> 0.9 (27°C) <sup>e</sup>
Coastal	76,000		
Wetlands	90,000		
Crops	110,000	<i>Frigoribacterium</i> sp. <sup>e, i</sup>	
Land ice	(5,000)	<i>Raphidonema</i> spp. <sup>j</sup>	$1.7 \cdot 10^{-4} - 2.91 \cdot 10^{-4}$ (12-18°C) <sup>j</sup>

Deserts	(10,000)		
	612 <sup>c</sup>		
Forests	56,000		
	6,323-12,748 <sup>d</sup>		
Shrubs	350,000		
Seas	10,000	<i>Pseudoalteromonas</i> <sup>k</sup>	0.25 (T unknown) <sup>k</sup>
Seas (estuary) <sup>b</sup>		Gamma-Proteobacterium (the fastest) <sup>l</sup>	0.17 (14°C) <sup>l</sup>
			0.19 (24°C) <sup>l</sup>

<sup>a</sup> All cell concentrations are taken from Burrows et al. (2009b) unless otherwise noted. <sup>b</sup> ‘seas estuary’ was not included as a separate ecosystem by Burrows et al. (2009b); <sup>c</sup> (Lighthart and Shaffer, 1994); <sup>d</sup> (Helin et al., 2017); <sup>e</sup> Amato et al. (unpublished data; strains originally reported in Amato et al., 2007c ~~or~~ and Vaitilingom et al., 2012); <sup>f</sup> (Männistö and Häggblom, 2006); <sup>g</sup> (Bakermans et al., 2003); <sup>h</sup>; (Morris et al., 2000) <sup>i</sup> (Copeland et al., 2015); <sup>j</sup> (Stibal and Elster, 2005); <sup>k</sup> (Middelboe, 2000) ; <sup>l</sup> (Fuchs et al., 2000)

We assume in **Section 3** that all bacteria cells as listed in **Table 1** are metabolically active. The atmospheric lifetime of bacteria cells is limited to several minutes (Otero Fernandez et al., 2019) to hours (Amato et al., 2015). In our estimates, we neither include assumptions on the limited lifetime of bacteria cells nor on their residence time in the atmosphere as it is assumed that PBA emissions lead to a continuous replenishment of bacteria in the atmosphere resulting in a steady-state concentration of living cells. The consequences of limited cell life- and residence time on SBA formation warrant further studies in more sophisticated model approaches.

## 2.2 Cell generation rates $R_{Cell}$

Different levels of metabolic activity can be distinguished, from survival, where cells only repair molecular damages, to maintenance (dormancy), where cells do not divide but maintain biological functions, to growth, allowing the net production of biological mass (Price and Sowers, 2004) (**Figure 1**). The generation rate of a microorganism during growth is probably the most common microbiological criterion used for characterizing microbial multiplication in the laboratory; it corresponds to the time that is needed for doubling the cell number, i.e., for producing two “children” cells from one individual. This requires mass production from nutrients that provide the necessary molecular bricks and biochemical energy. The activity depends on physiological traits of the microorganism, with optima at a given temperature, pH, salinity, and other conditions that define its fitness for its habitat. The generation time in bacteria at their optimum growth conditions usually ranges from ~20 minutes (Marr, 1991) to several days or weeks; as conditions deviate from the optima, this lengthen to virtually infinite time in non-dividing cells.

The *cellular* growth rate itself, i.e. the increase of individual cell size and mass, is intimately linked with generation time: cell size increases in a predictable way as generation time decreases (Si et al., 2017), and it can vary ~~of~~ **by** a factor of up to eight within a single bacteria species. Compared to generation rates, cellular growth rates are usually small and, thus, in the following only data for generation rates are used to estimate SBA mass formation rates.

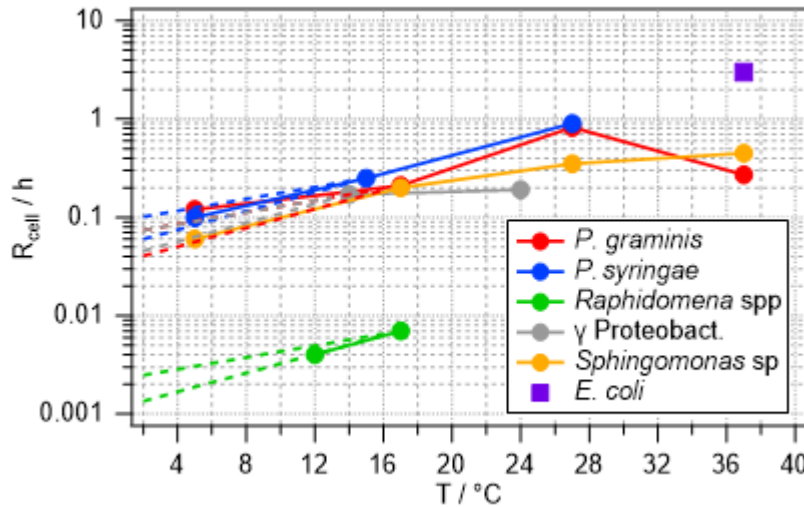
Metabolic activity, in terms of carbon uptake per units of biomass and time, can range over more than ten orders of magnitude, depending on many factors of which temperature is a major one (Price and Sowers, 2004). Therefore, if

available, temperature-dependent generation rates  $R_{cell}$  are listed in **Table 1** and shown in **Figure 2**. In addition, the highest expectable growth rate for bacteria as measured under laboratory conditions in culture medium is also shown in the figure for constraining an upper theoretical limit. This corresponds to the generation rate of the laboratory model *Escherichia coli* under optimal conditions (37°C). However, it can be expected that this is not representative of situations encountered in clouds.

Generally, the temperature dependence of cell generation rates can be scaled by the empirical relationship in **Eq-1**:

$$R_{cell}(T_2) = R_{cell}(T_1) \cdot Q_{10}^{(T_2-T_1)/10} \quad (1)$$

whereas  $R(T_1)$  and  $R(T_2)$  are the growth rates ( $h^{-1}$ ) at two temperatures  $T_1$  and  $T_2$ .  $Q_{10}$  is a dimensionless scaling factor that expresses the change of these rates over an interval of 10°C and typically has values between two and three within relatively small temperature intervals (Lipson et al., 2002; Sand-Jensen et al., 2007). In general, Equation (1) can be applied for all bacteria types and is usually valid for liquid water over a temperature range up to ~ 25°C; however, the slope ( $Q_{10}$  factor) and the maximum temperature depends on the bacteria type. In **Figure 2**, the dashed lines towards lower temperatures represent extrapolations of the generation rates at ~20°C reflecting the general agreement between



**Figure 2:** Temperature dependence of generation rates  $R_{cell}$  for bacteria types representative for the ecosystems in **Table 1** and *E. coli* as a likely upper limit for cell generation. Dashed lines are extrapolations of the rates at  $T \sim 20^\circ\text{C}$  using  $Q_{10} = 2$  or  $Q_{10} = 3$ , respectively. measured and calculated temperature dependencies using  $Q_{10} = 2$  or 3, respectively. Using generation rates measured at ~20°C might lead to an overestimate for SBA mass formation rates in colder clouds. However, we chose these values for the calculations in Section 3 as most experimentally-derived growth rates are available for temperatures of ~ 20 – 30°C. Generally, at temperatures below 0°C, cell metabolic activity is negligible in terms of carbon flux even though cells can maintain and survive under such conditions (Amato et al., 2009, 2010; Price and Sowers, 2004).

## 2.2 Bacteria growth efficiency (BGE)

Chemoheterotrophs - representatives of which were shown to maintain metabolic activity in clouds (Amato et al., 2017) - take up carbon from dissolved organic material for both recovering biochemical energy and converting the substrates into CO<sub>2</sub> and other products. Bacteria growth efficiency (BGE) is defined as the biological mass that is produced relatively to the amount of carbon taken up from the environment, the rest being converted into CO<sub>2</sub> (Eiler et al., 2003):

$$\text{BGE} = \frac{d[\text{organic products}]}{d[\text{organic products}] + d[\text{CO}_2]} \quad (2)$$

Note that in the original literature BGE is defined as a measure of ‘biomass production’ instead of organic products’ in Eq (2). Given the large body of atmospheric literature on aerosol processes that discusses ‘biomass’ as material from any living matter (e.g., aerosol from forest fires), we avoid using ‘biomass’ in the current context of microbial processes. BGE for planktonic bacteria range from < 0.4% to 80% with the highest values for eutrophic conditions (Eiler et al., 2003). In turn, in the same study, it was shown that, when substrate availability is limited, values from ~7% to ~14% are generally observed. As the conditions in cloud water can be considered oligotrophic with typical concentrations of dissolved organic carbon (DOC) of less than 0.1 mM (Herckes et al., 2013), low BGEs in the range of 0.1 – 10% can be expected, i.e. DOC is efficiently converted into CO<sub>2</sub>.

## 2.4 Cloud properties relevant for microbial activity

### 2.4.1 Cell concentrations in cloud water

Bacteria cells have sizes of up to several micrometers which explains their high efficiency to act as cloud condensation nuclei (CCN) (Bauer et al., 2003; Després et al., 2012). Assessing the hygroscopicity of biological particles is complex since it cannot be calculated in a similar way as for chemical compounds where total hygroscopicity represents the sum of the contributions of all components (Ariya et al., 2009). Once particles form cloud droplets, chemical compounds dissolved in cloud water, will trigger the growth of the processed CCN and, thus, enhancing hygroscopicity and CCN activity of aged particles in subsequent cloud cycles. The dissolution of ambient cell populations of 100 to 50,000 m<sup>-3</sup> (**Table 1**) results in 200 to 500,000 cells mL<sup>-1</sup> for clouds with liquid water contents (LWC) of 0.5 g m<sup>-3</sup> and 0.1 g m<sup>-3</sup>. The reasonable agreement of cell concentrations outside of clouds and those in cloud water suggests that a large fraction of bacteria cells are scavenged and, thus, act as CCN.

Some bacteria are well-known to efficiently act as ice nuclei (Amato et al., 2015; Möhler et al., 2008; Morris et al., 2004). In the current study, we neglect the potential role of ice clouds as media of microbial metabolic activity. In addition to low temperatures resulting in very low generation rates (**Figure 2**), the substrate diffusion to the bacteria will be limited resulting in negligible consumption of dissolved carbon.

## 2.4.2 Time fractions of microbial processes in clouds ( $F_{cloud}$ )

As we assume that both SBA mass production and WSOC loss only occur when bacteria are suspended in cloud droplets, we need to estimate the time bacteria spend in liquid clouds. In general, cloud contact times, i.e., the time air spends in a cloud, are dependent on cloud depth and vertical velocity (Feingold et al., 2013). This small-scale information is not consistently available for the large regions as covered by the ecosystems listed in **Table 1**. ~~Instead, the fraction of cloud contact time  $F_{cloud}$  is here equaled to the atmospheric volume fraction that is filled with clouds (Lelieveld and Crutzen, 1990).~~

~~Using global cloud coverage data based on satellite images derived from MODIS data (King et al., 2013) and an average cloud thickness of 1.5 km within the lowest 6 km of the atmosphere (Pruppacher and Jaenicke, 1995; Wang et al., 2000), we apply a factor of 0.25 (1.5 km / 6 km) to obtain an average cloud volume, and thus  $F_{cloud}$ , above each ecosystem. In order to give an estimate of the cloud processing time over the various large ecosystems as identified by Olson et al. (1992), we use the approximate cloud fractions during spring averaged for 2000 – 2011 from MODIS Terra (e.g., Figure 2b in (King et al., 2013)). This representation gives only a general view of cloudiness that varies over smaller spatial and temporal scales. However, given the conceptual nature of our study that builds upon the categories as used in the previous study by Burrows et al. (2009) for primary bacteria emissions, our approach seems sufficient to give (i) a reasonable estimate of cloudiness above the various ecosystems and (ii) enough detail of its concept to be refined in future studies.~~

Globally, a range of cloud thicknesses of 1.4 – 1.9 km has been derived (Table 1 in (Wang et al., 2000)) from which we use  $h = 1.5$  km as a single value for the average cloud thickness. Assuming further that globally > 90% of all liquid clouds reside in the lowest 6 km of the atmosphere ( $\Delta z = 6$  km) (Pruppacher and Jaenicke, 1995), we can convert the cloud coverage as obtained from satellite data ( $F_{clc}$  in Table 2) into cloud volume fractions using Eq-3:

$$F_{cloud} = F_{clc} \cdot \frac{\text{cloud thickness [km]}}{\Delta z \text{ [km]}} = F_{clc} \cdot \frac{1.5 \text{ km}}{6 \text{ km}} \quad (3)$$

Comparison of previous estimates of global cloud coverage of 60% (Pruppacher and Jaenicke, 1995) and the volume fraction of liquid clouds within the atmosphere of 15% (Lelieveld and Crutzen, 1990) generally supports this relationship. The resulting  $F_{cloud}$  values are summarized in **Table 2** together with the percentage area fraction of each ecosystem of the Earth surface, taken from Burrows et al. (2009b) and originally obtained from Olson (1992) and the cloud coverage data ( $F_{clc}$ ).

**Table 02:** Surface coverage of ecosystems on Earth surface (Burrows et al., 2009b), approximate cloud coverage  $F_{clc}$  above the ecosystems, estimated based on maps of annual cloud cover data obtained by MODIS, and estimated time fraction bacteria spend in clouds ( $F_{cloud}$ )

	<u>% of Earth surface <sup>a)</sup></u>	<u><math>F_{clc}</math></u>	<u><math>F_{cloud}</math></u>
--	---	-----------------------------	-------------------------------



<u>All</u>	<u>100</u>	<u>0.6</u>	<u>0.15</u>
<u>Tundra</u>	<u>3.3</u>	<u>0.4</u>	<u>0.1</u>
<u>Grassland</u>	<u>2.2</u>	<u>0.7</u>	<u>0.2</u>
<u>Coastal</u>	<u>0.2</u>	<u>0.4</u>	<u>0.1</u>
<u>Wetlands</u>	<u>0.6</u>	<u>0.5</u>	<u>0.15</u>
<u>Crops</u>	<u>3.0</u>	<u>0.7</u>	<u>0.3</u>
<u>Land ice</u>	<u>3.1</u>	<u>0.4</u>	<u>0.1</u>
<u>Deserts</u>	<u>3.7</u>	<u>0.2</u>	<u>0.05</u>
<u>Forests</u>	<u>7.0</u>	<u>0.9</u>	<u>0.25</u>
<u>Shrubs</u>	<u>5.8</u>	<u>0.3</u>	<u>0.1</u>
<u>Seas</u>	<u>71.0</u>	<u>0.7</u>	<u>0.2</u>

<sup>a)</sup> Data from Burrows et al. (2009b) and Olson (1992)

215

### 3. Results and Discussion

#### 3.1 SBA mass production

##### 3.1.1 Calculation of SBA formation rates

We calculate the SBA mass formation rate [ $\text{ng m}^{-3} \text{day}^{-1}$ ] above each ecosystem  $i$ , using Eq (34):

$$\left(\frac{dm}{dt}\right)_{\text{SBA},i,\text{day}} = R_{\text{cell},i} \cdot F_{\text{live}} \cdot C_{\text{cell},i} \cdot F_{\text{cloud},i} \cdot m_{\text{cell}} \quad (34)$$

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where  $R_{\text{cell}}$  is the cell generation rate [ $\text{h}^{-1}$ ] (**Table 1**). For ecosystems, for which  $R_{\text{cell}}$  of the representative bacteria types is not available, we assume the average formation rate of  $R_{\text{cell}} = 0.3 \text{ h}^{-1}$  as an upper limit for atmospherically-relevant conditions, corresponding to a generation time of approximately three hours.  $C_{\text{cell}}$  denotes the ambient cell concentration [ $\text{cell m}^{-3}$ ] (**Table 1**),  $F_{\text{live}}$  is the fraction of living cells in total cell concentration and assumed to be unity here,  $F_{\text{cloud}}$  is the fraction of total time ~~bacteria~~ when bacteria are active in clouds (**Table 2**), and  $m_{\text{cell}}$  is the average mass of a single cell, independent of the bacteria type. The cell mass  $m_{\text{cell}}$  is assumed to be  $52 \cdot 10^{-15} \text{ g cell}^{-1}$  (Sattler et al., 2001), equivalent of a spherical particle with diameter of 500 nm and a density of  $1 \text{ g cm}^{-3}$ .

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For nearly all ecosystems, predicted SBA formation rates are in the range of  $\sim 0.1$  to  $\sim 1 \text{ ng m}^{-3} \text{ s}^{-1}$  (**Figure 3a**), with higher values for crops and shrubs where  $C_{\text{cell}}$  were found to be highest (**Table 1**). The average value ( $0.6 \text{ ng m}^{-3} \text{ day}^{-1}$ ), calculated using the average values representative for all ecosystems (Category ‘All’ in Table 1), is similar to most of the ~~formation rates in the individual ecosystems~~ other formation rates, suggesting that using these average data for a global estimate gives a reasonable order of magnitude of SBA formation. Only above land ice where  $C_{\text{cell}}$  is small, the rate is significantly smaller. Given that the temperatures above land ice might be on average lower than above other regions, the relative importance of SBA formation there might be even smaller. According to the definition of the categories as suggested by Olson et al. (1992), the category ‘land ice’ does not include sea ice. It can be expected

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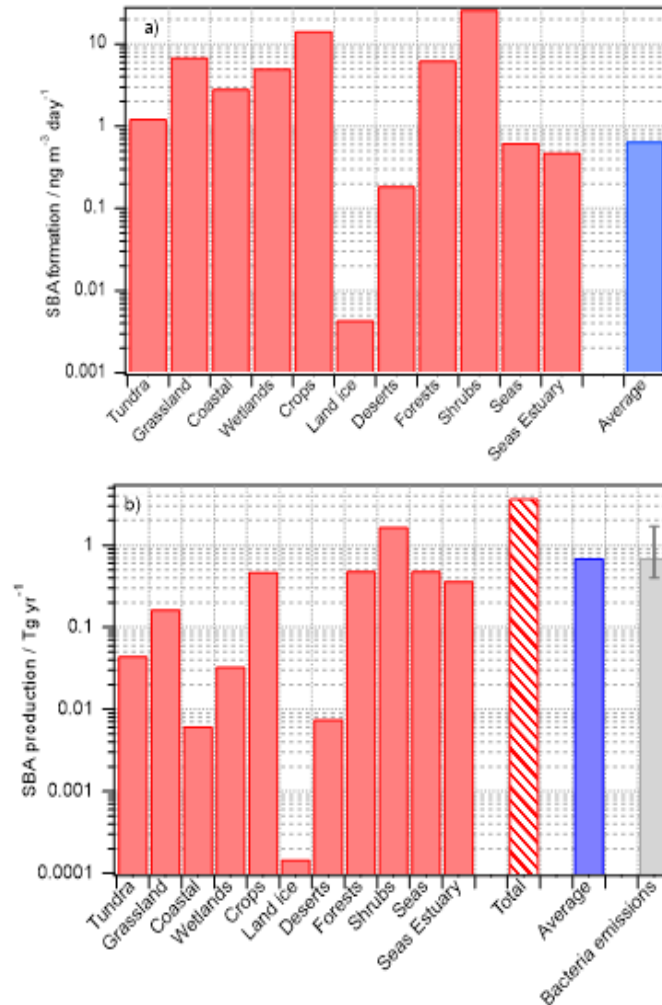
that above sea ice the sources and metabolic activity of bacteria are also very low (Martin et al., 2009) and thus can be likely neglected on a global scale.

To compare the mass production to other global aerosol sources,  $m_{SBA,day}$  is converted into a production flux [Tg yr<sup>-1</sup>] for each ecosystem  $i$  and scaled by the surface fraction of each ecosystem  $A_i$ :

$$P_{SBA,i} = \left( \frac{dm}{dt} \right)_{SBA,i,day} \cdot 365 \text{ days} \cdot A_i \cdot V_{atmos} \quad (45)$$

where  $V_{atmos}$  is the volume of atmosphere ( $3 \cdot 10^{18} \text{ m}^3$ ) and  $A_i$  is the surface fraction of each ecosystem (Table 2).

The production fluxes for each ecosystem are shown in Figure 3b, together with their sum for all ecosystems. The



**Figure 3:** Predicted production of secondary biological aerosol mass above the various ecosystems. The blue bar indicates the predicted production using the average values for all ecosystems ('all' in Table 1); a) SBA formation rates [ $\text{ng m}^{-3} \text{ day}^{-1}$ ]; b) SBA production rates [ $\text{Tg year}^{-1}$ ]. The red shaded bar represents the total predicted amount of SBA production is  $3.7 \text{ Tg yr}^{-1}$  with highest contributions from bacterial activities above seas ( $0.5 \text{ Tg yr}^{-1}$ ) as they cover most of the globe (71%) and above shrubs ( $0.5 \text{ Tg yr}^{-1}$ ) since the highest bacteria

245 concentrations have been identified there (*Table 1*). Forests with a much smaller surface area (7%) but higher cell  
concentration contribute the same amount (0.5 Tg yr<sup>-1</sup>). Using average data instead of those for individual ecosystems  
results in 0.7 Tg yr<sup>-1</sup>. Given the large uncertainties in all factors of Eq (34) and Eq (45), we suggest that this average  
(0.7 Tg yr<sup>-1</sup>) and the value based on the weighted sum of all ecosystems (3.7 Tg yr<sup>-1</sup>) might be considered a  
250 reasonable range of 'first best estimate' of total SBA contribution by bacteria on a global scale. These values are similar  
to the range of 1 - 10 Tg yr<sup>-1</sup> that was extrapolated by Sattler et al. (2001) based on carbon production rates of bacteria  
in supercooled clouds at mount Sonnblick observatory in the Austrian Alps. Our estimated SBA mass production  
represents the production of total bacteria mass. The carbon content of bacteria cells is roughly 50% of their dry mass,  
with the remainder composed of nitrogen, oxygen, phosphorous, hydrogen and other elements (Whitman et al., 1998).  
Thus, we suggest that SBA formation may lead to ~0.4—1.9 Tg carbon yr<sup>-1</sup> based on our best estimate that is bound  
255 in biological mass.

### 3.1.2 Discussion of uncertainties in SBA formation

The formation rates in *Section 3.1.1* represent an estimate of a previously unrecognized source of biological aerosol  
mass in the atmosphere. All parameters are associated with large uncertainties that need to be constrained in the future  
as they might vary depending on temporal, meteorological, spatial and geographical conditions. Ranges of observed  
260 values for all parameters in Eq (34) and Eq (45) are summarized in *Table 3* and discussed in the following:

(i) The cell concentrations  $C_{cell}$  in the atmosphere homogenize aloft due to mixing processes and average to  
concentrations of ~10,000<sup>4</sup> m<sup>-3</sup> at most locations. However, spatial deviations might be present in particular locations,  
such as cell concentrations of ~7·10<sup>5</sup> - 4·10<sup>6</sup> cells m<sup>-3</sup> >10<sup>6</sup> m<sup>-3</sup> that were found in highly polluted areas during haze  
periods in Chinaas (Li et al., 2018; Xie et al., 2018), and even 10<sup>9</sup> m<sup>-3</sup> above a wastewater storage lagoon which can  
265 be considered the highest expectable value of ambient bacteria near the ground (Paez-Rubio et al., 2005). Using the  
framework presented in the present study, SBA formation in such rather spatially limited areas can be estimated if  
growth rates of the individual bacteria types were available.

(ii) The growth rates  $R_{cell}$  assumed here likely represent an overestimate as cloud temperatures are likely often lower  
than ~15-20°C. At temperatures > ~0°C, this overestimate is likely less than an order of magnitude (*Figure 2*); in  
270 supercooled cloud droplets (< 0°C), metabolic activity  $R_{cell}$  might be some orders of magnitude lower and cell  
multiplication can be considered negligible.

(iii) While some of the studies listed by Burrows et al. (2009b) and in *Table 1* report the concentrations of viable cells,  
others give the total cell concentrations. In addition, the large discrepancy in reported  $F_{live}$  between < 0.1% up to nearly  
100% as discussed e.g., by Lindeman et al. (1982), Lighthart and Shaffer (1994) and Gandolfi et al. (2013), might be  
275 also due to differences in the measurement techniques. Consistent experimental methodologies are needed to give  
comprehensive data on  $F_{live}$  and the survival rates of bacteria in aerosol particles (Otero Fernandez et al., 2019).

(iv) The average cell mass depends on bacteria type and their growth stage. Sattler (2001) estimated carbon mass of  
bacteria cells in cloud water as 17 fg carbon cell<sup>-1</sup> in agreement with values for marine and freshwater ecosystems.

Approximating the total mass by doubling the carbon mass, results in  $34 \text{ fg cell}^{-1}$ , i.e. equivalent to spherical particles with diameters of  $\sim 0.4 \text{ }\mu\text{m}$ . Carbon mass and the carbon-to-total mass ratio can greatly differ from these values; for example, total masses of prokaryotic cell of  $200 \text{ fg cell}^{-1}$  in soil have been reported (Whitman et al., 1998). In their global study, Burrows et al. (2009b) assumed a mass of  $520 \text{ fg cell}^{-1}$  ( $1 \text{ }\mu\text{m}$  particle).

**Table 03:** Parameters used in the estimate of SBA mass formation and their possible minimum and maximum values based on literature data

Parameter	Value in Eq (34) and Eq (45)	Range		Comment
		Minimum	Maximum	
$C_{\text{cell}} [\text{m}^{-3}]$	$10^4$	100	$10^9$	$C_{\text{cell},\text{min}}$ : above desert during low RH and high radiation (Lighthart and Shaffer, 1994); $\sim 10^6$ : in a highly polluted area (Xi'an, China) (Xie et al., 2018); $C_{\text{cell},\text{max}}$ was measured <del>near the ground</del> <u>above a wastewater storage lagoon</u> (Paez-Rubio et al., 2005)
$R_{\text{cell}} [\text{h}^{-1}]$	0.3	0	3	$R_{\text{cell}} \sim 0$ might occur under stressful conditions when cells become dormant (e.g. low temperature, little water...); $R_{\text{cell}} \sim 3$ for <i>E. coli</i> under optimal conditions ( $37^\circ\text{C}$ , appropriate culture medium).
$F_{\text{live}}$	1	0.0001	1	0.0001 – 0.2 based on global microbial diversity (Gandolfi et al., 2013); 0.22 above crop fields (Lindemann et al., 1982), 0.81 above desert (Lighthart and Shaffer, 1994). Some studies report concentrations of viable cells; in this case reported concentrations imply $F_{\text{live}} = 1$
$m_{\text{cell}} [\text{fg cell}^{-1}]$	52	34	520	$m_{\text{cell},\text{min}}$ corresponds to cells in clouds assuming that <del>they</del> <u>cells</u> are composed of 50% carbon (Sattler et al., 2001); masses of other prokaryotic cells might be <u>up to</u> <del>200</del> $\text{fg cell}^{-1}$ (Whitman et al., 1998); $m_{\text{cell},\text{max}}$ <del>is</del> <u>for</u> <del>corresponding to</del> a spherical cell of a diameter $1 \mu\text{m}$ and density of $1 \text{ g cm}^{-3}$ (Burrows et al., 2009b).
$F_{\text{cloud}}$	0.15	$> 0$	1	The average global value might be higher than 0.15 if bacterial processes also occur outside of clouds. On small scales or above individual ecosystems, the value can be smaller than the average value, <u>depending on cloud variability</u> .

(v) While several studies have shown that liquid water is necessary for efficient microbial activity, it is not clear yet whether bacteria maintain activity in wet aerosols. Klein et al. (2016) found indications that bacteria metabolic activity exists in aerosols but no quantitative data is reported yet. Bacteria become dormant (Kaprelyants and Kell, 1993) or have reduced viability at relative humidities of 86 - 97% (Haddrell and Thomas, 2017). In soil samples, it has been shown that cycles of drying and rewetting might enhance microbial activity compared to constantly moist samples (Meisner et al., 2017; Xiang et al., 2008); thus, it may be speculated that such effects also occur in rapidly changing humidity conditions in atmospheric deliquesced aerosols. Under those conditions, the time fraction of microbial activity would exceed  $F_{cloud}$ . On locally smaller scales, cloud processing time might be shorter than the average  $F_{cloud}$  (Table 2) depending on cloud variability.

SBA mass formation calculated by Eq (34) and Eq (45) depends linearly on all parameters discussed in (i) to (v). Thus, the sensitivity-uncertainty of the predicted formation rates can be simply estimated by the ranges given in **Table 3**. However, in the atmosphere, all parameters might continuously change over time and thus might affect SBA mass to different extents.

### 3.1.3 Comparison of SBA formation to other aerosol sources

An estimate of aerosol emissions from the biosphere suggested a source strength of primary biological particles of 1000 Tg yr<sup>-1</sup> (Jaenicke, 2005). However, in this study, PBA was defined to include all cellular material, proteins, and their fragments. A global model study predicted total PBA emissions (bacteria, fungal spores and pollen) of 123 Tg yr<sup>-1</sup> of which bacteria comprised 0.79 Tg yr<sup>-1</sup> (Myriokefalitakis et al., 2017). This number is similar to the range of 0.4 – 1.8 Tg bacteria yr<sup>-1</sup> as given by Burrows et al (2009b). Bacteria usually comprise only a small fraction of total PBA; a major fraction is composed of fungal spores. Thus, their emissions are generally estimated to be larger than those of bacteria. For example, Fungal spores are considered a major fraction of PBA. Their global emissions (25 Tg yr<sup>-1</sup>) was suggested to contribute 23% to total primary organic aerosol (Heald and Spracklen, 2009). An estimate of fungal spore emissions based on tracer compounds resulted in predicted 50 Tg yr<sup>-1</sup> (Elbert et al., 2007)). None of these estimates include microbial activity as a source of biological mass. Our predicted SBA source of 0.7–3.7 Tg year<sup>-1</sup> is on the same order of magnitude as similar to the model predictions for primary bacteria emissions. The estimates of primary bacteria emissions are performed such that observed cell concentrations are matched by the models without considering another source of cells in the atmosphere. Our SBA estimates might be equally biased as they are based on the same ambient cell concentrations as used-reported by Burrows et al. (2009a, b) and Myriokefalitakis et al. (2017). The absolute values and the ratio of primary to secondary bacteria mass need to be evaluated by more complex model studies as our simple framework can provide.

Total organic aerosol is composed of mostly secondary mass. Best estimates based on observational and model studies of the net production rate of secondary organic aerosol (SOA) mass are on the order of 136 – 280 Tg year<sup>-1</sup> (Hodzic et al., 2016). These amounts are similar to the predicted global sulfate production of 117 Tg year<sup>-1</sup> (39 Tg S year<sup>-1</sup>)

(Chin et al., 2000). Thus, SBA production can be estimated to be on the order of ~1% of the ~~total~~-secondary aerosol sources. The net aerosol mass formation due to SBA production might be even smaller if bacteria metabolize substrates that are already in the particle phase. In this case, biotransformation processes lead to the conversion of non-biological into biological aerosol mass. The unique properties of biological aerosol material have been extensively discussed in the context of heterogeneous ice nucleation where it has been shown that even small amounts of biological material could have significant effects on clouds and precipitation (Möhler et al., 2008; Morris et al., 2004; Santl-Temkiv et al., 2015). Given the low ambient concentrations of ice nucleating particles and their high sensitivity to the ice/liquid partitioning in mixed-phase clouds (e.g., Ervens et al., 2011), a small change in biological mass possibly translates into significant changes in the evolution of cold clouds.

### 3.2 Consumption of organic carbon in clouds

#### 3.2.1 Calculation of microbial and chemical WSOC loss rates

Bacteria can be metabolically active in the aqueous phase of clouds (Delort et al., 2010) and on the surface or bulk phase of particles (Klein et al., 2016; Estillore et al., 2016). Such processes lead to a decrease of water-soluble organic carbon (WSOC) mass within cloud droplets as bacteria convert substrates into CO<sub>2</sub> (**Figure 1**). Also processes of biological mass production from CO<sub>2</sub> exist (autotrophy) and include photosynthesis (photoautotrophs). However, despite the fact that photosynthetic microorganisms were reported in the atmosphere (Tesson and Santl-Temkiv, 2018) there is neither a clear indication yet of photosynthetic activity in clouds, nor of other modes of autotrophy.

The split between carbon uptake for biological mass production and mineralization is quantified by the bacterial growth efficiency BGE (Eq (2)). Studies have shown that generally metabolic processes produce mostly CO<sub>2</sub> under atmospheric conditions and only a small fraction of the carbon is mineralized into organic products (< 1 – 10% of the total C utilized). We introduce here the factor  $F_{CO_2}$  as a measure of the loss of organics due to bacterial processes:

$$F_{CO_2} = 1 - BGE \quad (56)$$

Using  $F_{CO_2}$ , we can write the carbon balance as

$$WSOC \xrightarrow{\text{Bacteria}} F_{CO_2} CO_2 + (1 - F_{CO_2}) WSOC \quad (R1)$$

The loss rate of carbon can be, thus, calculated as

$$R_{WSOC,Bact} \left[ \frac{g}{L(aq) s} \right] = - \frac{d(WSOC)}{dt} = \frac{dCO_2}{dt} = \quad (67)$$

$$-F_{CO_2} k_{Bact} \left[ \frac{1}{cell \cdot s} \right] \cdot C_{Cell,aq} \left[ \frac{cell}{L} \right] \cdot F_C \cdot C_{WSOC} \left[ \frac{gC}{L} \right] \cdot LWC \left[ \frac{g}{m^3} \right] \cdot F_{cloud} -$$

$$-F_{CO_2} k_{Bact} \left[ \frac{L(aq)}{cell \cdot s} \right] \cdot C_{Cell,aq} \left[ \frac{cell}{L(aq)} \right] \cdot F_C \cdot C_{WSOC} \left[ \frac{gC}{L(aq)} \right] \cdot F_{cloud}$$

350 whereas the cell concentration in cloud water can be replaced by

$$C_{Cell,aq} \left[ \frac{\text{cell}}{\text{L(aq)}} \right] = C_{cell,g} \left[ \frac{\text{cell}}{\text{m}^3(\text{g})} \right] / \text{LWC} \left[ \frac{\text{m}^3(\text{g})}{\text{L(aq)}} \right] \quad (8)$$

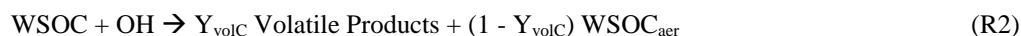
~~where with~~  $C_{Cell,aq}$  and  $C_{WSOC}$  ~~are being~~ the concentrations of bacteria and water-soluble organic carbon in cloud water, respectively, and  $C_{cell,g}$  ambient cell concentrations in the gas phase (e.g. Table 1). These concentrations are on average  $\sim 10^7$  cells  $\text{L}^{-1}$  (Vařtilingom et al., 2013) and 0.1 mM (Herckes et al., 2013) in cloud water whereas both values might differ over a few orders of magnitude locally and temporally (**Section 3.2.2**). Usually experimental loss rates of organics by bacteria in real and artificial cloud water are reported to be on the order of  $\leq 10^{-17}$  mol  $\text{cell}^{-1}$   $\text{s}^{-1}$  for organic substrates (e.g., formic, acetic, and succinic acids) (Vařtilingom et al., 2010, 2011). The cell activity is dependent on the bacteria type and the availability of the organic substrate. Thus, strictly, such rates [mol  $\text{cell}^{-1}$   $\text{s}^{-1}$ ] are only valid for the substrate-to-cell ratio as applied in the experiments. In order to account for the ratio as encountered in cloud water, we use here  $k_{Bact}$  [ $\text{L cell}^{-1}$   $\text{s}^{-1}$ ] (i.e. measured rate divided by the concentration of organic substrate in the experiments) that is applicable to the full range of conditions where the cells exhibit a similar microbial activity. Resulting rate constants for formic, acetic and succinate acids are on the order of  $k_{Bact} \sim 10^{-13}$   $\text{L cell}^{-1}$   $\text{s}^{-1}$ . Equation 7 includes an additional factor  $F_C$  in Eq (5) that accounts for the microbial selectivity towards only some organics by each bacteria type (e.g. Šantl-Temkiv et al., 2013; Bianco et al., 2019). ~~Bacteria are selective towards organic compounds and not all organics are metabolized by every bacteria type.~~ For example, it has been shown in a single study that, upon laboratory incubation of cloud water, oxalic acid is not affected by cloud borne microorganisms, formate is only consumed after a lag time of several hours, which is much longer than the lifetime of a cloud droplet, and, on the other hand, compounds such as acetate or succinate are readily biodegraded (Vařtilingom et al., 2011). Since these compounds comprise major constituents of WSOC in cloud water, it seems reasonable to introduce a factor  $F_C < 1$ . ~~an additional factor  $F_C$  in Eq (5) that accounts for the microbial selectivity towards only some organics by each bacteria type (e.g. Šantl-Temkiv et al., 2013; Bianco et al., 2019).~~ Given the complexity of the organic matter in the atmosphere, the numerous organic molecules existing in cloud water and their variable susceptibility to biodegradation,  $F_C$  is hard to specify with precision. Bianco et al. (2019a) observed experimentally by FT-ICR-MS during laboratory incubation of cloud water that out of the 2178 compounds detected, 1094 were actually utilized by bacteria (~50%). Assuming that all these compounds were equally abundant, one could conclude that 50% of all cloud water organics were prone to be microbiologically consumed (i.e.  $F_C = 0.5$ ). More quantitative support of this assumption could be given based on the fact that preferably small oxygenated organics are taken up by bacteria. Compilations of speciated cloud water organics have shown that small acids, such as formic and acetic acid, comprise a large fraction (at least ~30%) of the characterized fraction of cloud water organics (e.g. Figure 6 in Herckes et al., (2013). Lab experiments have shown that these acids are degraded by bacteria (Amato et al., 2007a; Vařtilingom et al., 2013). ~~Obviously~~ While  $F_C = 0.5$  seems a reasonable compromise, this factor is highly uncertain and strongly depends on the microbial and chemical composition of cloud water. The calculated rates  $R_{Bact,WSOC}$  are summarized in **Figure 4** for  $0.8 \leq F_{CO2} \leq 0.99$  and  $F_C = 0.5$ .

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385 Several studies have discussed the competition of microbial and chemical processes in cloud water as a sink of specific organic compounds (Ariya et al., 2002; Husárová et al., 2011; Vaithilingom et al., 2010, 2013). The most efficient loss reactions for organics in cloud water are initiated by OH radicals. The general rate constant of the OH radical with water-soluble organic carbon is  $k_{OH} = 3.8 \cdot 10^8 \text{ M}^{-1} \text{ s}^{-1}$  (Arakaki et al., 2013). The reactions of WSOC with OH lead to volatile and non-volatile oxidation products. Radicals are much less selective towards organics than bacteria are; thus, the assumption of a factor equivalent to  $F_C$  in Eq (67) is not necessary as all water-soluble organics react with OH whereas the chemical reactivity mostly depends on the structure of the organic compound. The yield of volatile products ( $Y_{volC}$ ) includes  $\text{CO}_2$  but also formaldehyde and other volatile compounds that do not remain in the particle phase after cloud evaporation and thus do not contribute to the aerosol loading. We assume  $0.2 \leq Y_{volC} \leq 0.5$ , but -in general,  $Y_{volC}$  depends on the WSOC composition, with higher values for more aged organics that are more readily oxidized to volatile products.

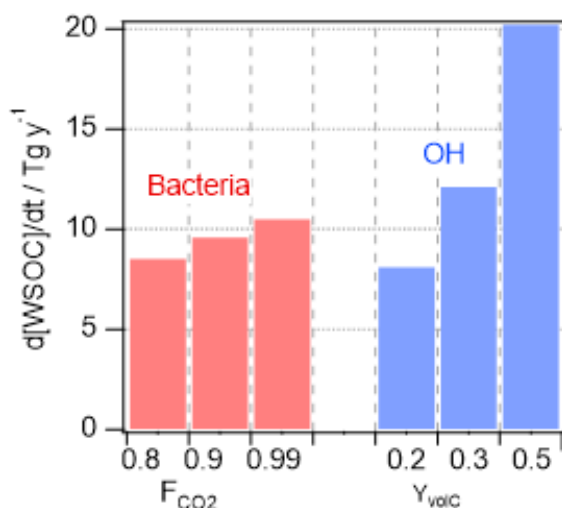
395 Equivalent to reaction (R 1), we express the carbon loss by the OH radical in clouds as



With  $\text{WSOC}_{aer}$  the WSOC fraction that remains in the aerosol phase after drop evaporation. We calculate the loss rate accordingly:

$$\begin{aligned}
 R_{OH,WSOC} &= - \frac{d(\text{WSOC})_{OH}}{dt} \left[ \frac{\text{g}}{\text{L(aq)s}} \right] \\
 &= k_{OH} [\text{L mol}^{-1} \text{ s}^{-1}] [\text{OH}]_{aq} [\text{mol L}^{-1}] \cdot Y_{volC} C_{WSOC} \left[ \frac{\text{gC}}{\text{L}} \right] \cdot \text{LWC} \left[ \frac{\text{g}}{\text{m}^3} \right] \cdot F_{cloud} \quad (79)
 \end{aligned}$$

OH concentrations in cloud water are in the range of  $10^{-16} \text{ M} < [\text{OH}]_{aq} < 10^{-14} \text{ M}$  (Arakaki et al., 2013; Bianco et al., 2015; Ervens et al., 2014) and an average cloud liquid water content (LWC) of  $0.15 \text{ g m}^{-3}$  is assumed. The results in



**Figure 4:** Predicted loss of WSOC by bacterial utilization and by chemical (OH) processing in cloud water for different assumption on  $F_{CO_2}$  and  $Y_{volC}$ ,

*Figure 4* suggest that the microbial rates may be smaller than the chemical ones under most conditions. Overall, the values shown in *Figure 4* only differ by a factor of  $\sim 2.5$  which might imply that there are conditions under which chemical and biological processes in the aqueous phase compete. show that the predicted rates  $R_{OH,WSOC}$  ( $8 - 20 \text{ Tg yr}^{-1}$ ) and  $R_{Bact,WSOC}$  ( $8 - 11 \text{ Tg yr}^{-1}$ ) are for most parameter sets on a similar order of magnitude with possibly a slightly higher contribution of chemical reactions to WSOC loss. This trend is in agreement with several previous studies that focused on the comparison of microbial versus chemical processes as sinks for specific organic substrates (Amato et al., 2007a; Vaitilingom et al., 2010). These loss fluxes are relatively large as compared to the predicted SBA formation (*Figure 3*).

In a previous study, it was estimated that microbial processes in clouds lead to a total carbon loss of  $\sim 10 - 50 \text{ Tg yr}^{-1}$  and to a production of  $\sim 100 \text{ Tg year}^{-1} \text{ CO}_2$  with the assumptions of complete respiration ( $F_{CO_2} = 1$ ), microbial non-selectivity towards WSOC ( $F_C = 1$ ) and applying the same loss rates as observed in lab experiments without correcting for differences in the ratio of bacteria cell to WSOC concentrations (Vaitilingom et al., 2013). Thus, this former estimate can be considered an upper limit whereas the one in the current study ( $\sim 50 - 30 \text{ Tg yr}^{-1}$ ) is more conservative, both suggesting that the respiration of bacteria is a negligible  $\text{CO}_2$  source as compared to the sum of anthropogenic sources ( $\sim 50,000 \text{ Tg CO}_2 \text{ year}^{-1}$  (IPCC, 2014)).

The WSOC loss rates calculated here are also much smaller than those estimated for the loss of total SOA due to heterogeneous reactions and photolysis on particle surfaces ( $\leq 50.3 \text{ Tg year}^{-1}$ ) and by wet deposition ( $\leq \sim 50 \text{ Tg year}^{-1}$ , (Hodzic et al., 2016)). For the parameters chosen in our estimate, they are also smaller than the predicted production rates of aqSOA in clouds of  $13.1 - 46.8 \text{ Tg year}^{-1}$  (Lin et al., 2014) or  $20 - 30 \text{ Tg yr}^{-1}$  (Liu et al., 2012). This estimate represents organic carbon sources and sinks in general. It should be noted that the organics comprising WSOC and aqSOA, respectively, might not be identical. These predictions strongly depend on the representation of clouds, and particularly on the liquid water content (He et al., 2013), but overall emphasize that clouds can be considered a net source rather than a sink for organic aerosol.

### 3.2.2 Discussion of uncertainties of microbial and chemical WSOC loss

The calculation of microbial and chemical WSOC loss should be regarded an approximation using a set of parameters that are all associated with considerable uncertainties. Similar to the discussion in *Section 3.1.2*, we compile all parameters and minimum and maximum values based on literature data in *Table 4*:

(i) There are not as many measurements of  $C_{Bact,aq}$  as for cell concentrations  $C_{cell}$  in cloud-free regions. The assumption that all particles that contain bacteria cells are activated into cloud droplets does likely not lead to a large overestimate. The sizes of bacteria-containing particles usually exceed several hundred nanometers and thus can all be considered CCN. Differences in LWC as the conversion factor from gas to aqueous phase concentrations are relatively small, within a factor of 2 – 3, within the categories of common cloud types (Pruppacher and Klett, 2003).

(ii) The activity of microorganisms towards organic substrates is often reported in units of ‘mol(substrate) cell<sup>-1</sup> s<sup>-1</sup>’ which expresses the amount of substrate that is consumed per cell and time. For several compounds (e.g., formate, acetate, succinate) these rates differ by approximately one order of magnitude (Vaitilingom et al., 2011). However,

the resulting  $k_{Bact}$  values (i.e. rate divided by substrate concentration) are all on the order of  $k_{Bact} \sim 10^{-13} \text{ L cell}^{-1} \text{ s}^{-1}$  which appears to represent an upper limit for the organics that have been investigated for metabolic activity in clouds.

440 A much lower constant was derived from experiments with less oxygenated compounds such as phenol (Lallement, 2017).

While we only consider the direct interaction of bacteria and organics, additional processes might lead to more complex chemical and microbial interactions. For example, siderophores form iron complexes (Passananti et al., 2016) and, thus, suppress Fenton reactions that affect oxidant levels in cloud droplets (e.g., Deguillaume et al., 2004). Such  
445 indirect feedbacks of microbial processes on chemical budgets require more comprehensive data sets that are currently not available for models.

(iii) The respiration of bacteria depends on many different factors such as stress due harsh conditions. It can be expected that at higher stress levels (nutritional ~~ef~~or thermal),  $F_{CO_2}$  increases to supply elevated energy needs (Amato and Christner, 2009; Eiler et al., 2003). Values of BGE as low as  $< 0.4\%$  ( $F_{CO_2} = 0.996$ ) were observed (Eiler et al.,  
450 2003 and references therein), indicating that nearly all the carbon used was mineralized into  $CO_2$ .

(iv) The fraction of organic material metabolized by bacteria in clouds ( $F_C$ ) is likely not unity for a single bacteria type (Bianco et al., 2019; Vaitilingom et al., 2011). Carboxylic acids that are preferentially metabolized by several common bacteria types often comprise a major fraction ( $\sim 20\%$ ) of the cloud-water organics that can be speciated on a molecular level (Herckes et al., 2013). This fraction might be regarded a lower limit of  $F_C$  since the reactivity of the  
455 large fraction of unspciated organics (often  $\sim 70\%$ ) towards bacteria is not known. However, a recent qualitative study suggested that  $\sim 50\%$  of all organics in cloud water are microbially consumed by bacteria (Bianco et al., 2019). Our comparison implies the same spatial accessibility of bacteria and OH, respectively, to WSOC. This might be an oversimplification as bacteria are unevenly distributed among cloud drop populations as statistically only one in  $\sim 10,000$  droplets may contain a single bacteria cell. OH can be expected to be present in all cloud droplets as the direct  
460 phase transfer from the gas phase represents one of the major OH sources in cloud water.

**Table 4:** Values for parameters in Eq (67) and Eq (79) used in the estimate of WSOC loss by microbial and chemical processes and their likely minimum and maximum values

Parameter	Value in Eq (67) and Eq (79)	Range		Comment
		Minimum	Maximum	
$C_{Bact,aq}$ [cells $L^{-1}$ ]	$10^7$	$10^6$	$10^8$	Range of total bacteria concentration observed in cloud water samples collected from a mid-altitude mountain site over several years (31 samples) (Vaitilingom et al., 2012)
LWC [g $m^{-3}$ ]	0.15	0.1	1	The minimum and maximum value describe a range for a wide variety of cloud types. The assumption of LWC is not needed if it is assumed that all bacteria-containing particles act as CCN.

$k_{\text{Bact}}$ [L cell <sup>-1</sup> s <sup>-1</sup> ]	10 <sup>-13</sup>	10 <sup>-15</sup>	10 <sup>-13</sup>	$k_{\text{Bact,min}}$ was derived for microbial activity towards phenol (Lallement, 2017). $k_{\text{Bact,max}}$ was derived from experiments using cloud water and is valid for the microbial activity of various highly oxygenated compounds.
$F_{\text{CO}_2}$	0.8 – 0.99	0.2	1	Even though BGE ranging from <-0.4% to 80% (0.996 > $F_{\text{CO}_2} > 0.2$ ) were estimated in natural environments (Eiler et al. 2003 and references therein), at low nutrient concentrations, as encountered in clouds, high $F_{\text{CO}_2}$ can be expected.
$F_{\text{C}}$	0.5	0.2	< 1	Herckes et al. (2013) report that ~20% of total organic carbon in clouds is composed of speciated carboxylic acids; <a href="#">Bianco et al. (2019) demonstrate that ~50% of all organics in cloud water are affected by bacteria.</a> $F_{\text{C}} = 1$ seems unlikely due to variation in microbial and chemical cloud water composition.
$k_{\text{OH}}$ [L mol <sup>-1</sup> s <sup>-1</sup> ]	3.8·10 <sup>8</sup>	10 <sup>6</sup>	10 <sup>10</sup>	Typically, undissociated acids (low pH) and polyfunctional compounds have $k_{\text{OH}}$ at the lower end of this range whereas the upper limit is constrained by diffusion limitation (Herrmann, 2003; Monod and Doussin, 2008)
[OH] <sub>aq</sub> [M]	10 <sup>-15</sup>	10 <sup>-17</sup>	10 <sup>-14</sup>	The suggested range includes concentrations that were inferred for night-time conditions (minimum) to day-time conditions in clean air masses (low OH sinks).
$Y_{\text{volC}}$	0.3 – 0.5	0.2	0.8	This value has not been comprehensively quantified yet; largest values can be likely expected in aged WSOC with high O/C ratios

(v) While the absolute importance of microbial loss depends on the parameters discussed in (i) to (iv), the relative importance compared to chemical processes might be of interest in studies where the fate of individual organics in the cloud droplets or in the atmospheric multiphase system is explored. [OH]<sub>aq</sub> depends mostly on photochemical processes as source processes and on the concentrations of WSOC as the main sinks; it ranges from 10<sup>-17</sup> M (night-time) to 10<sup>-14</sup> M (day time, clean air masses) (Arakaki et al., 2013).

(vi) Given that formate and acetate comprise major contributors to cloud water organics (Herckes et al., 2013), some fraction of WSOC will be converted into highly volatile products, such as CO<sub>2</sub> and CH<sub>3</sub>CHO that will not remain in the particle phase after cloud evaporation. However,  $Y_{\text{volC}}$  likely does not approach unity since several studies have suggested that radical reactions in cloud water lead to the successive decay of dicarboxylic acids into their next smaller

homologue which will remain in the aqueous phase. Within these limits, we conservatively suggest a range of 0.2 to 0.8 for  $Y_{volC}$  but point out the need for studies to refine this parameter

#### 475 4. Summary and conclusions

We have estimated the amount of biological mass that is formed in the atmosphere by growth and multiplication of bacteria cells ('secondary biological aerosol', SBA). Data for representative bacteria strains and their generation rates have been compiled for major ecosystems. Using average values for cloudiness above the various ecosystems, we estimate that 3.7 Tg year<sup>-1</sup> SBA mass is formed globally which is comparable to current estimates of direct bacteria emissions (0.4 – 0.7 Tg yr<sup>-1</sup> (Burrows et al., 2009b; Myriokefalitakis et al., 2017)) which comprise a small fraction of total biological aerosol mass. While these production rates make up ~1% of other major secondary aerosol formation rates (secondary organics or sulfate), their importance might differ on spatial or temporal scales. In addition, SBA production leads to an increase in biological aerosol mass which might sensitively affect physicochemical particle properties (e.g. ice nucleation ability). SBA formation linearly depends on several parameters, such as the number concentration of metabolically active bacteria cells, their generation rates and the time scales during which they are assumed to grow or multiply – all of which are associated with considerable uncertainties. The ecosystem categories in Table 2 represent fairly large regions. It might be expected that SBA formation rates are different on smaller spatial and/or temporal scales. For example, it has been shown that human activities in cities lead to high bacteria concentrations; also forests have been identified as significant sources of biogenic aerosol. However, detailed data on bacteria are sparse in such regions. While several recent studies have characterized the diversity of microorganisms in forested regions (rainforest, tropics) (Gusareva et al., 2019; Souza et al., 2019), these studies did not report cell concentrations which highlights the urgent need of additional measurements.

~~Our~~ The detailed discussion of the parameters and their uncertainties in our simplified approach highlights the likely variability of SBA formation on smaller scales and the need of future studies to refine these parameters. Similar approaches as ours may be applied to yeast growth. Yeast cells are generally larger (~2 – 10 μm) than bacteria cells (Fröhlich-Nowoisky et al., 2009) and, thus, their residence time in the atmosphere is likely shorter. Detailed data on their activity in clouds are not available which currently prevents the assessment of their potential contribution to SBA.

We also quantify the role of clouds as sinks of total WSOC by microbial and chemical processes, unlike previous studies that focused on microbial activity towards individual organic compounds. It is estimated that microbial processes lead to an organic mass loss of 8 – 11 Tg yr<sup>-1</sup> whereas chemical processes by the OH radical in clouds lead to a loss of 8 – 20 Tg yr<sup>-1</sup>. These numbers are small compared to other sinks such as aerosol removal by deposition. Not all of the organic-WSOC mass even contributes to organic aerosol loading as water-soluble, volatile organics are dissolved in cloud water but evaporate during drop evaporation. Thus, the loss of organic aerosol mass due to direct microbial activity in clouds is-might be smaller than the predicted loss of organic-carbonWSOC. Large uncertainties in these estimates represent the assumptions on the fraction of carbon that is converted into volatile products. For

bacteria, this fraction is quantified by the bacteria growth efficiency that depends on numerous factors, such as bacteria type, substrate availability and physical conditions in the condensed phase.

510 In current atmospheric models, when considered, bacteria cells are inert, i.e. they neither change their mass or number concentrations during their residence time in the atmosphere nor do they interact with other aerosol constituents. The approach presented in our study ~~is~~ provides a first simplified estimate of SBA formation and WSOC loss due to bacteria that could be easily adapted in models. Given the current great activities in the field of atmospheric bioaerosols, it can be expected that the discussed parameters in the estimates can and should be refined in the future in order to quantify the role of bacterial processes as source of biological mass and net source or sink of organic  
515 aerosol in the atmosphere.

**Data availability.** All data are available upon request from the authors.

**Author contributions.** BE and PA planned and carried out the study and wrote the manuscript together.

**Competing interests.** The authors declare that they have no conflict of interest.

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