A point by point answer to reviewers for the article

To Reviewer RC1

Thank you so much for the detailed review work to improve the clarity and understanding of the paper. Responses to all your comments can be found below, together with the amended paper containing the track changes.

- **Line 43**: suggest replace "shall" with "should"

  --> **done**
- **Lines 44-47**: This abstract text is repetitive, and I recommend it be cut.

  --> **We have cut the sentence**

  "The H&S parameterisation shows satisfactory results over northern and eastern France, but may underestimate concentrations for Mediterranean areas that may indicate missing factors influencing emissions or a missing source of spores."
- **Line 51**: Inappropriate references for the text. The two references use AMS observations which measure particles less than ~1 um, not PM10. Pai et al. do not show an underestimate of in situ observations.

  --> **We have replaced the sentence** *"Modelling of the organic matter (OM) fraction of PM10 chronically underestimates in situ observations (Ciarelli et al., 2016; Pai et al., 2020). This underestimation can be attributed to several causes such as the complexity of the organic matter composition, which is not yet fully known, incomplete emission inventories or their inherent uncertainties, and poorly parametrised atmospheric chemical transformations."* **by** *"Modelling of the organic matter (OM) fraction of PM10 with chemical transport models can be complex due to the varied composition of organic matter, which is not yet fully known, incomplete emission inventories or their inherent uncertainties, and poorly parameterised atmospheric chemical transformations."*
- **Line 90**: typo? Remove "the variability"

  --> **It was a typo. We have changed the sentence**

  "*amaké et al. (2019a) identified the parameters responsible the variability for up to 82 % of the annual variability of polyols […]***"**

  by "*Samaké et al. (2019a) identified the parameters responsible for up to 82 % of the annual variability of polyols […]***"**
- **Lines 113-114**: The number of data points seems like excessive detail that bogs down the introduction.

  --> **We have replaced the sentence** *"This comparison was carried out using 1,536 hourly data points, 1,200 of which came from the German and Finnish stations, each with 600 data points."*

by "*This comparison was carried out using 1,536 hourly data points, that most of which came from the German (600) and Finnish (600) stations.***"**

- **Lines 179-182**: PMF is also commonly used for source apportionment of online measurements (such as AMS). Suggest you re-phrase.

  --> **We have replaced the sentence** *"Positive Matrix Factorisation (PMF) is one of the most widely used techniques for identifying factors contributing to aerosol concentrations using field measurements"*

  by "*Positive Matrix Factorisation (PMF) is one of the most widely used techniques for identifying factors contributing to aerosol concentrations using online and offline measurement data***"**
- **Table 1**: Provide units or more details in caption for the column "PMF" and "Polyols". Are these number of samples?

  --> **We have replaced the caption** *"Summary of organic matter and polyols filter-based observations as well as primary biogenic factor […]"*

  by "*Summary of the number of daily filters analysed for polyols as well as OM from primary biogenic factor […]*"
- **Line 231**: The reference Fountoukis and Nenes (2007) refers to ISORROPIA II – either correct the text to refer to this version or cite the original reference for ISORROPIA I.

  --> **We have replaced the text "***The ISORROPIA thermodynamic model […]***" by "***The ISORROPIA II thermodynamic model […]***"**
- **Section 2.1**: should also describe the simulation of other PM10 components (e.g. BC, dust, sea salt) since PM10 simulations are included in the manuscript.

  --> **A description of the simulation of the chemical composition of PM10 and its evaluation is provided in a separate paper. For spores, no interaction with other species was taken into account. This is why we have chosen not to go into detail on the simulation of all the other species.**
- **Table 2**: This provides extraneous or duplicative information from the text. Suggest cut.

  --> **We have cut the Table 2**
- **Line 271**: reference for this sentence?

  --> **We have cut the sentence** *"This yields to large emissions especially over Southern Europe which are not confirmed by measurements."*
- **Equation 1**: why are there multiple constants (c, 5, 1.5x10-2) rather than integrating these into a single constant?

  --> **We have rewritten the equation from Hoose et al. (2010). We have added the simplified version** *"30 867 × LAI × qv".*
- **Line 291**: please include units for density.

--> **We have modified the sentence** *"Fungal spore number concentrations are transformed into mass using an aerosol density of 1 […]"*

by *"Fungal spore number concentrations are transformed into mass using an aerosol relative density of 1 kg kg-1 […]"*

- **Lines 290-292**: What size of fungal spore is assumed for this conversion? Are the spores treated as a size distribution or monodisperse?

  --> We have added the sentence "*Fungal spores are treated as non-soluble particles and considered as monodispersed with a diameter of 3 µm in a model size class closest to 3 µm. In the model configuration with 10 size bins, spores are included in the size bin 8 corresponding to sizes between 2.5 and 5 µm, with an average diameter of 3.5 µm.*"
- **Line 298**: are the fungal spores treated as soluble?

  --> No, this is now mentioned in the sentence added in response to the previous comment.
- **Section 3.1** discussion of lifetime: Lifetime depends on the assumed size and solubility of the aerosol. These model assumptions should be described (see previous comments) and the lifetime diagnosed for the specific simulation. In particular, line 341 "Despite little transport" has not been shown. Are there any spores aloft in the simulation? Any transported to the eastern boundary of the domain?

  --> There are indeed concentrations of spores at higher altitudes (> 20 metres) but obviously much lower than in the first 20 metres. On the other hand, the transport at the eastern boundary of the domain is negligible. As mentioned in the text, deposition is fairly rapid, given the large size of the particles. Hummel et al (2015) calculated a lifetime of about 5h for biogenic fluorescent aerosols (FBAP). If we compare the simulated fungal spore emission and concentration maps, we see a very strong similarity indicating a short lifetime.

  We have added the sentence *"Since only the transport and deposition of fungal spores are taken into account, without interactions with other species, the conditions are similar to those of Hummel et al. (2015), with a lifetime of the same order of magnitude (5 hours), further confirming the low transport of spores."* **and changed the sentence** *"Despite little transport"* **by** *"Despite these conditions of transport and deposition"*
- **Line 445**: It seems odd to end the discussion of Figure 4 without discussing the outliers Aix and Nice. This comes later in the text, so the authors may wish to explicitly tell the reader that they will discuss these outliers in Section 3.2.2, or move this discussion forward.

  --> We have added the sentence "*The complicated results observed at stations on the Mediterranean coast (Aix-en-Provence, Marseille, Nice, Port-de-Bouc) will be discussed in the next section."*
- **Line 460**: why do the authors use MFE here when using MFB in Figure 4. I suggest harmonizing for consistent statistical metrics.

  --> Both statistical metrics are different and used in parallel. We have replaced the sentence *"Finally, for comparison with the same polyol data set, daily and*

monthly mean fractional error (MFE) are respectively 0.79 and 0.56 at all sites"

by "*Finally, for comparison with the same polyol data set, daily and monthly mean fractional bias (MFB) are respectively -11 % and -11 % at all sites (Table S1, Figure S1). The root mean square error (RMSE) and the mean fractional error (MFE) was also calculated for estimating the error (Table S1, Figure S2). Daily and monthly MFE are respectively 79 % and 56 % at all sites; for the median RMSE the results are respectively 0.04 µg m^{-3} and 0.03 µg m^{-3}."*

- **Figure 5** and **lines 539-546**: Please include all sites with long-term measurements discussed in the text in Figure 5.

  --> We have included in Figure 5 all the timeseries for non-Mediterranean sites. The timeseries for OMpb and polyols for all sites are in the supplement.

To Reviewer RC2

Thank you so much for your careful review. I am honoured that you found the study interesting and that the paper is for you well written and easy to follow. Responses to all your comments can be found below, together with the amended paper containing the track changes.

- **Line 172**: Out of 699 stations in France, fungal spore tracers are observed in 13. If the full station network is not used to evaluate model results in the present study, I find it confusing to read about the 699 stations in the beginning.

  --> **Initially this paper included an assessment of PM10 and we forgot to delete this sentence. In the end, it was moved to a separate paper that will be submitted in the next few days. We have removed the sentence** *"A total of 699 air quality stations performed measurements in metropolitan France during the period of the study, restricted to 2013 and 2014, including fixed and mobile stations."*
- **Line 188**: Are these 7 sites from the 13 sites in total used in this study? And why only these 7?

  --> As mentioned in Table 1, the sites with OM data for the primary biogenic PMF factor are among those with polyol measurements. However, not all sites were subject to a PMF study since the complete chemical composition was not carried out. This explains why not all the sites measuring polyols have PMF data. The same applies to the number of PMF vs. polyol data, which can be explained by the fact that the chemical composition of PM10 was not carried out on all the filters.
- **Lines 190-195**: is there an estimate of how significant this contribution can be?

  --> Yes, there are estimations, and we modified the following sentence to include one. We have replaced the sentence *"PMF results at all sites include a factor which can be attributed to PBOA because of the large concentrations of the two polyols in this factor, representing more than 90 % of them."*

  by "*PMF results at all sites include a factor which can be attributed to PBOA because of the large concentrations of the two polyols in this factor, representing more than 90 % of the polyols total mass in this factor (Samaké et al., 2019a).***"**
- **Table 1**: It would be great if you could provide the altitude information.

  --> We have included altitude information in Table 1.
- **Figure 1**: Is it possible to have the map showing the land use types and denote the measurement type, ie polyol vs PMF?

  --> We have included the number of data for each site in Figure 1 respectively for OMpb and polyols.
- **Line 243**: Could you comment on the 9x9 km spatial resolution representing the traffic stations.

--> The aim of this study is to look at the concentrations of primary biogenic aerosols. Since no interaction with other species is taken into account and these aerosols are emitted by vegetation, resolution at traffic sites is likely not an issue there. Furthermore, our results show good correlations and low biases at traffic sites for fungal spores. There could be an impact on resuspension, but this is not taken into account in the model for organic aerosols within the model.

- **Lines 274-276**: Would be good if you can show the statistics of these 3 different parameterizations (H&S, Hummel, and Janssen) in the supplement? It would be interesting to see the impact of considering wind as used in Janssen, in relation to the hypothesis in lines 323-325. The correlations may be close but how about the magnitudes?

  --> Thank you for this suggestion. As these results are already published in the literature and not part of our work (we rely on them), we refrain from reproducing them in our work, but they are easily accessible in the open scientific literature. Please note that the evaluation of the H&S and Hummel parametrisations are performed with the same biogenic fluorescent aerosol data (Hummel et al., 2015) and that the Janssen parametrisation has been evaluated alone in a separate paper on a spore dataset (Janssen et al., 2021). The differences in magnitudes can be highlighted with the discussion of biases.
- **Line 286**: So, the fungal spores are emitted in only one size (3 mm)? It would be good to discuss this assumption, rather than using a size distribution?

  --> We have added the sentence "*Fungal spores are treated as non-soluble particles and considered as monodispersed with a diameter of 3 µm in a model size class closest to 3 µm. In the model configuration with 10 size bins, spores are included in the size bin 8 corresponding to sizes between 2.5 and 5 µm, with an average diameter of 3.5 µm.*"
- **Line 330**: Could you infer the lifetime through your simulations? It would be interesting to compare with Hummel.

  --> Calculating a spore lifetime is complex. We only used the deposition and transport processes on the spores like in Hummel et al. (2015), so we assume that the spore lifetime should be similar. As mentioned in the text, deposition is fairly rapid, given the large size of the particles. Hummel et al (2015) calculated a lifetime of about 5h for biogenic fluorescent aerosols (FBAP). If we compare the simulated fungal spore emission and concentration maps, we see a very strong similarity indicating a short lifetime.
- **Line 435**: Does Chimere calculate OM or OC? If the latter, how is the OC converted to OM?

  --> Chimere simulates OM. In section 2.1.2 we have added the sentence "*The organic carbon of the primary biogenic PMF was multiplied by a factor of 1.8 to obtain the organic matter concentrations of this PMF factor (OMpb) (Favez et al., 2010; Petit et al., 2015).***"**
- **Figure 4**: Please explain in the caption what the box plot and the symbols represent.

  --> We have added the sentence *"Correlations and mean fractional bias (MFB) boxplots are obtained using the scores for each site (1 point per site) and are*

illustrated respectively at left and right side. The points corresponding to the sites represent the scores obtained on all the data for the site."

- The poorest model performance seems to be on the urban sites (traffic and industrial). Is this due to model resolution? Or the meteorological model not performing well for the urban areas?

  --> It's not a question of the type of site, but of the location. The most complicated sites to model are those close to the Mediterranean. This may be due to the resolution of the model, but above all to the different climate and possibly sources, which can have an impact on spore concentrations. This is already largely discussed in the paper and summarised at the end of the discussions section (section 3.3).
- **Lines 495-499**: Is this because of meteorology? Winds, precipitation, etc? It would be good to write in the methodology if these are nudged simulations. There might be an issue with transport, which could be identified by evaluating the model winds (speed and direction) that can explain the large underestimation in the southern sites?

  --> The meteorological data come from the WRF model, which was run in parallel with the CHIMERE model for the simulations and indeed nudged. The resolution of WRF meteorological data are consistent with the resolution chosen for the model, i.e. 9 x 9 km² in our study. For the 9 x 9 km² simulations, the domain and the simulations are not the result of nesting; the 9 x 9 km² European domain is our only domain and our only simulation. We have identified the meteorology as a source of problems in the Mediterranean area, with night-time temperatures thought to play a major role.

  We have replaced the sentence *"The WRF model is used for meteorological forcing (Skamarock et al., 2008)."*

  by *"The WRF 3.7.1 model (Skamarock et al., 2008) is used for meteorological simulation coupled to chimere with no aerosol effect. The spectral nudging (Von Storch et al., 2000) is used with NCEP temperature, wind, humidity, pressure for wavelengths higher than 2 000 km whereas in the boundary layer the WRF model freely generates its own dynamic."*