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Modelling of atmospheric concentrations of fungal spores: a two-year simulation over France using CHIMERE.

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Abstract

Fungal spore organic aerosol emissions have been recognised as a significant source of particulate matter as PM_{10}; however, they are not widely considered in current air quality models. In this work, we have implemented the parameterisation of fungal spore organic aerosol (OA) emissions introduced by Heald and Spracklen (2009) (H&S) and further modified by Hoose et al. (2010) in the CHIMERE regional chemistry-transport model. This simple parameterisation is based on two variables, leaf area index (LAI) and specific humidity. We have validated the geographical and temporal representativeness of this parameterisation on a large scale by using yearly polyol observations and primary biogenic organic aerosol factors from PMF analysis at 11 French measurement sites. For a group of sites in northern and eastern France, the seasonal variation of fungal spore emissions, displaying large summer and small winter values, is correctly depicted. However, the H&S parameterisation fails to capture fungal spore concentrations for a smaller group of Mediterranean sites with less data availability both in terms of absolute values as well as seasonal variability, leading to strong negative biases especially during the autumn and winter seasons occur. Two years of CHIMERE simulations with the H&S parameterisation have shown a significant contribution of fungal spore OA to PM_{10} mass, lower than 10 % during winter, and reaching up to 20 % during summer in high emission zones, especially over large forested areas. In terms of contribution to organic matter (OM) concentrations, the simulated fungal spore contribution in autumn is as high as 40 % and reaches at most 30 % of OM for other seasons. As a conclusion, the fungal spore OA contribution to total OM concentrations is shown to be substantial enough to be considered as a major PM_{10} fraction and shall then be included in state-of-the-art chemistry transport models. 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.

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1. Introduction

Modelling of the organic matter (OM) fraction of PM_{10} 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.

It is therefore important to assess whether the primary source of organic aerosol, currently not considered in many models, can help to improve atmospheric aerosol modelling. Primary biogenic organic aerosols (PBOA) are mainly composed of microorganisms such as bacteria, fungi, fungal or bacterial spores, pollens or viruses and biological fragments such as plant debris or microbes (Després et al., 2012; Fröhlich-Nowoisky et al., 2016; Jaenicke et al., 2007). Their size varies from less than 0.3 µm for viruses to about 100 µm for pollens (Després et al., 2012; Jones and Harrison, 2004; Shaffer and Lighthart, 1997). When looking at atmospheric particles with an aerodynamic diameter of less than 2.5 or 10 µm (which are the fractions routinely measured and studied for health risk assessment), it is possible to find viruses, bacteria (agglomerated or not) and spores; however spores, when produced by fungi, represent the major fraction in terms of mass (Elbert et al., 2007).

More specifically, fungal spores are emitted directly into the atmosphere during the fungal reproduction process when temperature and humidity conditions are favourable, but their emission can also be triggered by wind and rain (Elbert et al., 2007; Huffman et al., 2013; Jones and Harrison, 2004). Previous studies estimated that fungal spores can contribute to around 5 % and 10 % of the mass of respectively PM_{10} and organic carbon, in urban and suburban areas (Bauer et al., 2002, 2008b). In specific environments such as tropical forests, the contribution of fungal spores can represent 45 % of the PM_{10} mass (Elbert et al., 2007).

Fungal spores are susceptible to cause major health problems such as asthma, pulmonary obstruction, tuberculosis, meningitis and legionellosis (Douwes et al., 2003; Eduard et al., 2012; Fröhlich-Nowoisky et al., 2016; Ghosh et al., 2015; Pearson et al., 2015; Samaké et al., 2017). Some studies on PBOA have shown that aerosols emitted directly by fungi in the form of spores contribute significantly to the oxidative potential of aerosols (Samaké et al., 2017). Moreover, based on a positive matrix factorisation (PMF) analysis, Weber et al. (2021) derived a primary biogenic factor based on a large data set of speciated PM_{10} aerosol measurements over France, including polyol measurements as a tracer for fungal spores. They found a high intrinsic oxidative potential by dithiothreitol (DTT) for this factor, equal to that of biomass burning, but lower than that of primary traffic emissions.

Literature review shows several parameterisations suitable for use of modelling primary biogenic aerosols emissions from fungal spores in the PM_{10} size range in chemistry transport models. Samaké et al. (2019a) identified the parameters responsible the variability for up to 82 % of the annual variability of polyols as a tracer of fungal spores for a temperate latitude site in an alpine environment, using multi-linear approaches. These variations were mainly explained by the mean night-time temperature (54 %) and LAI (37 %), and to a lesser extent by the atmospheric humidity (3 %) and the wind speed (2 %). The combined factor of LAI and wind speed explains the remaining variability (4 %). A first parameterisation for the treatment of fungal

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scores in atmospheric models was proposed by Heald and Spracklen (2009) (H&S) and modified by Hoose et al. (2010). It estimates fungal spore emissions as a linear function of leaf area index (LAI) and specific humidity. In this formulation, the LAI is a proxy for the vegetation density and the specific humidity is a proxy for the water availability, but is also related temperature. The parameterisation proposed by Sesartic and Dallafior (2011) (S&D) suggests a different approach by varying emissions as a function of soil types, not relying on LAI, and therefore removing the seasonality inherently present in the H&S parameterisation. Hummel et al. (2015) compared these parameterisations across Europe and developed a new statistical model, based on the H&S parameterisation using LAI and specific humidity, to also include a linear dependence with temperature, and a threshold below which emissions are assumed to be zero.

In Hummel et al. (2015), the concentrations simulated with three parameterisations of H&S, S&D and Hummel were compared to measurements of fluorescent biological aerosol particles (FBAP) at four sites in several parts of Europe (Germany, Finland, UK, Ireland) for almost weekly time periods in July, August and October of 2010. 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. At these two sites, one week in July, one week in August and 10 days in September were measured, unlike the UK and Irish sites, where the data was taken only for August 2010. FBAP measurements are taken as a proxy for fungal spore emissions. By construction, the S&D parameterisation does not reproduce the observed daily and seasonal variability, while it is known that fungal spore emissions display a general summer maximum across Europe (Samaké et al., 2019a, b). On the contrary, the H&S and Hummel parameterisations include these temporal variations and therefore show better correlations with measured concentrations (R = 0.43) compared to the S&D approach (R = -0.05). The parameterisation by Hummel et al. (2015) showed a lower normalised mean bias (NMB = -43 %) compared to the H&S one (NMB = -44 %).

As fungal spores make a significant contribution to PM_{10} and are rarely included in chemistry transport models (CTM), the aim of our study is to integrate them into the state-of-the-art Chemistry Transport Model CHIMERE (Menut et al., 2021), to evaluate the model performance with field measurements, and to infer the spatio-temporal variability of their occurrence. This could lead to improved modelling of PM_{10} concentrations, of organic matter, and of other pollutants such as secondary biogenic compounds or even oxidative potential. This study will focus on France, displaying one of the largest database of chemically speciated PM measurements in Europe (Favez et al., 2021). Interestingly, France has a wide range of climatic variability (oceanic, semi-oceanic, continental, mountainous, Mediterranean), making it possible to compare fungal spore modelling results under various climatic conditions. To assess the modelling of fungal spores, measurements of polyols were used, specifically mannitol and arabitol, since many studies indicate that they are specific tracers of this PBOA fraction (Bauer et al., 2008a; Gosselin et al., 2016; Samaké et al., 2019a). Furthermore, we compared our CTM results to the concentrations of organic matter ascribed to this primary biogenic source using the receptor model Positive Matrix Factorisation (PMF) in previous work.

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2. Material & methods

2.1. Observations

2.1.1. PM_{10} and Organic matter measurements

The PM_{10} mass concentration data have been obtained from continuous measurements performed by French regional air quality monitoring networks (AQMN). 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. These observations have been achieved by AQMN using two types of automated analysers during this period: tapered element oscillating microbalances equipped with filter dynamic measurement systems (TEOM-FDMS, Thermo Scientific), and beta radiation absorption analysers (Met One BAM 1020 and ENVEA MP101M). These measurements have been conducted in accordance with standard procedures described in CSN EN 16450. As described by Favez et al. (2021), the aim of the aerosol characterisation program (CARA) is to develop knowledge of the chemical composition and contribution of atmospheric particle sources. This work is enriched by research programmes, with data from some of them being used in this study. In CARA and other programs, the chemical analysis of (PM_{10}) filter samples has been performed following relevant European standard methods. Briefly, for datasets used herein, organic carbon was measured by thermo-optical analysis using the EUSAAR2 protocol (Cavalli et al., 2010). Sugars were measured by liquid chromatography using pulsed amperometric detection (Verlhac et al., 2013; Yttri et al., 2015). The measurement protocols have been detailed in previous studies (Samaké et al., 2017, 2019a, b; Weber et al., 2021). The analysed species include mannitol and arabitol, which currently make up for a large fraction of organic sugars (Elbert et al., 2007) and are used as a tracer for fungal spore emissions.

In summary, for the datasets used in the present study, PM_{10} organic matter observations were performed at 13 different stations for a total of 2,227 daily filter samples, including 1,497 data on polyols on 11 sites. The locations of these sites are illustrated in Figure 1, while Table 1 provides details on the number of data points available per station and their temporality.

2.1.2. OC apportionment based on filter samples

Positive Matrix Factorisation (*PMF*) is one of the most widely used techniques for identifying factors contributing to aerosol concentrations using field measurements (Belis et al., 2020; Hopke et al., 2020; Karagulian et al., 2015; Paatero and Tapper, 1994). This receptor model commonly uses off-line chemical speciation measured on filters and factor-specific tracers as input data. The correlation matrices allow the identification of the species co-emitted with the tracers and thus determine the contribution of the factors to the PM_{10} concentrations. For this study, *PMF* analysis were previously performed with a harmonised methodology (Weber et al., 2021), providing source apportionment results for a total number of 842 daily samples collected at 7 sites from early 2013 to the end of 2014. 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. However, this PBOA factor may also contain biogenic secondary organic aerosols (BSOA) since it is sometimes associated with BSOA tracers, such as 3-MBTCA (resulting from α-pinene

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oxidation) or 2-MTs (resulting from oxidation of isoprene) (Borlaza et al., 2021). Therefore, we propose here to use the PBOA factor as an upper boundary for fungal spore concentrations (see section 3.2).

Table 1 : Summary of organic matter and polyols filter-based observations as well as primary biogenic factor derived from PMF analysis available for this study over the years 2013 and 2014 at different French sites (within the PM_{10} fraction). The measurement period and geographical coordinates are also indicated.

Stations	Coordinates (latitude ; longitude)	Measurement period	PMF	Polyols
Aix-en-Provence	43.53 ; 5.44	18.07.2013 – 13.07.2014	56	117
Andra-OPE	48.55 ; 5.46	01.01.2013 – 29.12.2014	/	98
Grenoble	45.16 ; 5.74	02.01.2013 – 29.12.2014	237	238
Lens	50.44 ; 2.83	05.04.2013 – 26.09.2014	167	138
Marseille	43.30 ; 5.39	01.06.2014 – 31.12.2014	/	95
Nice	43.70 ; 7.29	04.06.2014 – 31.12.2014	77	89
Nogent-sur-Oise	49.28 ; 2.48	02.01.2013 – 31.12.2014	155	220
Port-de-Bouc	43.40 ; 4.98	01.06.2014 – 31.12.2014	79	80
Revin	49.91 ; 4.63	02.01.2013 – 26.09.2014	/	168
Roubaix	50.71 ; 3.18	20.01.2013 – 08.09.2014	/	159
Strasbourg	48.59 ; 7.74	02.04.2013 – 31.12.2014	71	95

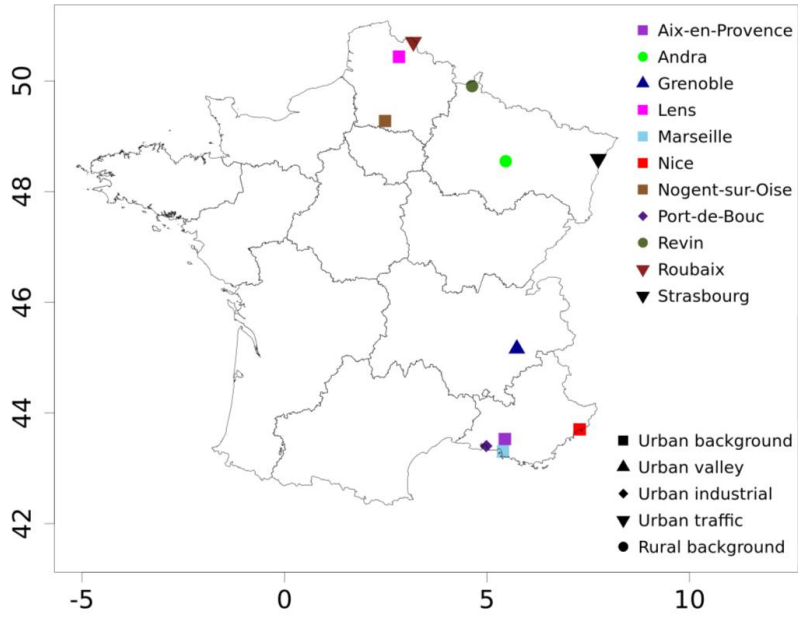

Figure 1 : Location and type of sites for PM_{10} organic matter and polyol measurements from filters as well as primary biogenic from PMF over the years 2013 and 2014.

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Sites are distributed over different geographical areas (Figure 1) in in the northeast and southeast of France, including cities from the Channel region (Lens, Roubaix, Nogent sur Oise) to the German border (Strasbourg), remote rural sites located in between (Revin and Andra-OPE) as well as sites an Alpine urban station (Grenoble) and sites near the Mediterranean Sea (Aix-en-Provence, Marseille, Nice, Port-de-Bouc). These sites are classified as rural background (Andra-OPE, Revin), urban background (Aix-en-Provence, Grenoble, Lens, Marseille, Nogent-sur-Oise, Nice, Petit Quevilly, Talence), traffic sites (Roubaix, Strasbourg), urban industrial (Port-de-Bouc). It is thought that the varied characteristics of the observational sites can give us an unprecedented possibility of evaluation of the simulated spore emissions and concentrations.

2.2. Regional Modelling

2.2.1. The chemistry transport model CHIMERE

CHIMERE is an eulerian state-of-the-art regional chemistry transport model (Menut et al., 2021). It is used operationally by the French platform PREV'AIR (Rouil et al., 2009) and the Copernicus Atmospheric Modelling System (CAMS) (Marécal et al., 2015) to forecast and monitor air quality. The version v2020r3 of CHIMERE has been used in this work (Menut et al., 2021).

The EMEP anthropogenic emissions inventory with a resolution of 10 km² provides input data for anthropogenic emissions based on the methodology described in Vestreng (2003). Biogenic VOC emissions are computed by CHIMERE based on the Model of Emissions and Gases and Aerosols from Nature MEGAN 2.1 algorithm (Guenther et al., 2012). The gas phase chemistry is provided by the Melchior2 mechanism (Derognat et al., 2003). The ISORROPIA thermodynamic model is used to compute the formation of inorganic aerosols based on the approach described in Fountoukis and Nenes (2007). For organic aerosol formation and volatilisation of primary organic aerosol, the volatility basis set (VBS) for the organic species as described in Cholakian et al. (2018) was activated.

Chemical boundary conditions with a 3-hour temporal resolution are from the CAMS project (Marécal et al., 2015), together with the chemical fields for the model upper boundary at the 500 hPa level. The WRF model is used for meteorological forcing (Skamarock et al., 2008). For the emissions of biogenic volatile organic compounds (VOC) as well as for the parameterisation of the emissions of primary organic aerosols, we use the LAI (Leaf Area Index) obtained from the observations of the MODIS instrument with a frequency of 8 hours and a native resolution of 30 seconds for each year (Sindelarova et al., 2014). The simulation has been carried out during years 2013 and 2014 on a western European domain, with a 9 x 9 km² horizontal resolution. It is run on 9 vertical hybrid levels from ground to an upper height of 500 hPa, the height of the first layer being around 20 meters.

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Table 2 : Parameterisations initially considered for the present work.

Name in this work	Variables	Reference
H&S	Leaf area index (LAI), specific humidity (q_v)	(Heald and Spracklen, 2009; Hoose et al., 2010)
S&D	Land use classes	(Sesartic and Dallafior, 2011)
Hummel	Leaf area index (LAI), specific humidity (q_v), surface temperature (T)	(Hummel et al., 2015)
Janssen statistical	Leaf area index (LAI), specific humidity (q_v), and wind friction speed (u*)	(Janssen et al., 2021)

2.2.2. Parameterisation of fungal spore OA emissions

In the Introduction section, we have presented several parameterisations of fungal spores, and which are listed in Table 2. Among the three of them (H&S, S&D and Hummel) compared by Hummel et al. (2015) to observations, the S&D parameterisation showed the worst statistical agreement, and also is based on seasonally fixed land-use parameters. It was therefore discarded. Among the two better performing parameterisations, we preferred the H&S parameterisation. This is because in Hummel's approach, the inclusion of a temperature-dependent and vegetation-independent term leads to significant fungal spore emissions under high temperature conditions even at places where LAI is small and therefore no large emissions are expected. This yields to large emissions especially over Southern Europe which are not confirmed by measurements. Finally, two recent parameterisations by Janssen et al. (2021) have been developed over the eastern United States using measurements of spore concentrations consider LAI, specific humidity and wind friction velocity in the first case, and a spore population model in the second. Comparisons with annual measurements of fluorescent primary organic aerosols at German, Finnish and Colorado sites show similar correlations between these two parameterisations and that of H&S (Janssen et al., 2021). With respect to these simulations, we preferred the simpler H&S parameterisation. This parameterisation was integrated in our simulations for its robustness at different sites and it has been set-up specifically for temperate latitude European conditions.

Equation 1 shows the fungal spore emission flux $\boldsymbol{F_{H\&S}}$ (unit: number of spores m^{-2} s^{-1}) varying as a function of leaf area index **LAI** and specific humidity $\boldsymbol{q_v}$. The constant $\boldsymbol{c}$, equal to 2315, introduced by Hoose et al. (2010) accounts for fungal spore emission fluxes with an aerodynamic diameter of 3 µm instead of 5 µm (which was initially estimated).

$$\boldsymbol{F_{H\&S}} = \boldsymbol{c}\frac{\boldsymbol{LAI}}{\boldsymbol{5}}\frac{\boldsymbol{q_v}}{\boldsymbol{1.5 \times 10^{-2}}} \quad (1)$$

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Fungal spore number concentrations are transformed into mass using an aerosol density of 1 which is used as reference density for the definition of aerodynamic diameter. All mass is attributed to organic matter. Within CHIMERE, fungal spores OA are prescribed as a new species considered as chemically inert in our simulation, but they can influence the condensation of semi-volatile secondary organic compounds (as part of the organic aerosol phase) and act as cloud condensation nuclei (Patade et al., 2021). However, no conclusive laboratory data are available to include such processes in a model. Other processes considered in the model apart from emissions are transport, and size-resolved dry and wet deposition with characteristics like that of primary anthropogenic aerosols.

3. Results

We will present here the results of the two-year long simulations containing fungal spores' organic aerosol. Our initial analysis delves into the variability of simulated emissions and concentration patterns, along with their impact on the simulated PM_{10} levels. We will then present an assessment of the simulated concentration fields with respect to polyol observations as well as primary biogenic organic burden as determined by the source apportionment studies.

3.1. Simulated two years of fungal spore primary organic aerosol

Figure 2 presents the seasonal variation in emissions and concentrations of fungal spore primary organic aerosols for the years 2013 and 2014 averaged, as well as that of LAI and specific humidity, obtained from our simulations. As parametrised, emissions are largely driven by vegetation density (represented here by the LAI) with emission structures that follow the distribution of the main French forest areas. Major forested areas and emission hotspots are seen over the Massif Central (centred at 2 °E, 45.5 °N), the Jura (6 °E, 47 °N), the lower parts of the Alps (7 °E, 46 °N) and Pyrenees (0 °E, 43 °N), and the Landes Forest (-1 °W, 45 °N). Specific humidity, which is the other parameter used explicitly in the flux calculation (equation 1), is more homogeneous and its signature on the fluxes of spore emissions is less easily identifiable. LAI and specific humidity show the same seasonal cycles with higher values in summer and lower values in winter when the vegetation density and water content of the colder atmosphere are lowest. We can therefore hypothesise that LAI and specific humidity are responsible for much greater fungal spore emissions in summer than in winter.

Concentrations of atmospheric spores are found to be highly correlated with emissions, both spatially and on a seasonal scale. Small differences can be explained by transport and deposition processes. For instance, due to advection, contrasts in concentrations are less pronounced than those in emissions. Hummel et al. (2015) assumes that the lifetime of fungal spores is of about 5 hours in the atmospheric boundary layer. This short lifetime means that there is a small chance of long-distance transport, which explains the closeness of local concentrations to emission sources. In our simulations, the total deposition flux of fungal spores can reach a maximum of

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10 ng m^{-2} s^{-1} on average over the two years, with 8 ng m^{-2} s^{-1} for dry deposition and 5 ng m^{-2} s^{-1} for wet deposition. In summer, this total spore deposition reaches a maximum of 20 ng m^{-2} s^{-1} in France, around 12 ng m^{-2} s^{-1} in the Massif Central, while spore emissions peak in this area at 25 ng m^{-2} s^{-1} on average over the summer period. The difference in emissions and deposition is therefore significant, confirming also partial transport out of source regions.

Despite little transport, spore concentrations at locations up to a few hundred kilometers away can be similar in mass and temporal variation, explained by similar meteorological conditions and leaf area index, leading to simultaneous emissions (Samaké et al., 2019b). Seasonal averages of fungal spore concentrations can reach values of several µg m^{-3} over large geographical areas, especially over the forested areas in the southern part of France (Massif Central). This is significant in view of the PM_{10} concentration there and consistent with previous studies (Heald and Spracklen, 2009). For instance, fungal spore OA contributes to about 20 % of PM_{10} mass on summer averaged over the Massif Central (Figure 3). On the contrary, during winter, fungal spore concentrations remain always below 0.5 µg m^{-3}, and do not contribute much to PM_{10}, with a value always below a few percent. Spring and autumn are intermediate, both in terms of fungal spore OA concentrations and contributions to PM_{10}. With the lower formation of BSOA compared to summer, the contribution of fungal spores to OM is largest in autumn, when it can reach around 40 %. It can reach about 30 % in other seasons with some geographical disparities. Despite low emissions and concentrations, the contributions of spores to concentrations of biogenic organic aerosols (BOA) is greatest in winter, reaching up to 70 %, due to the very low contribution of secondary biogenic organic aerosols during this period, in contrast to the summer period.

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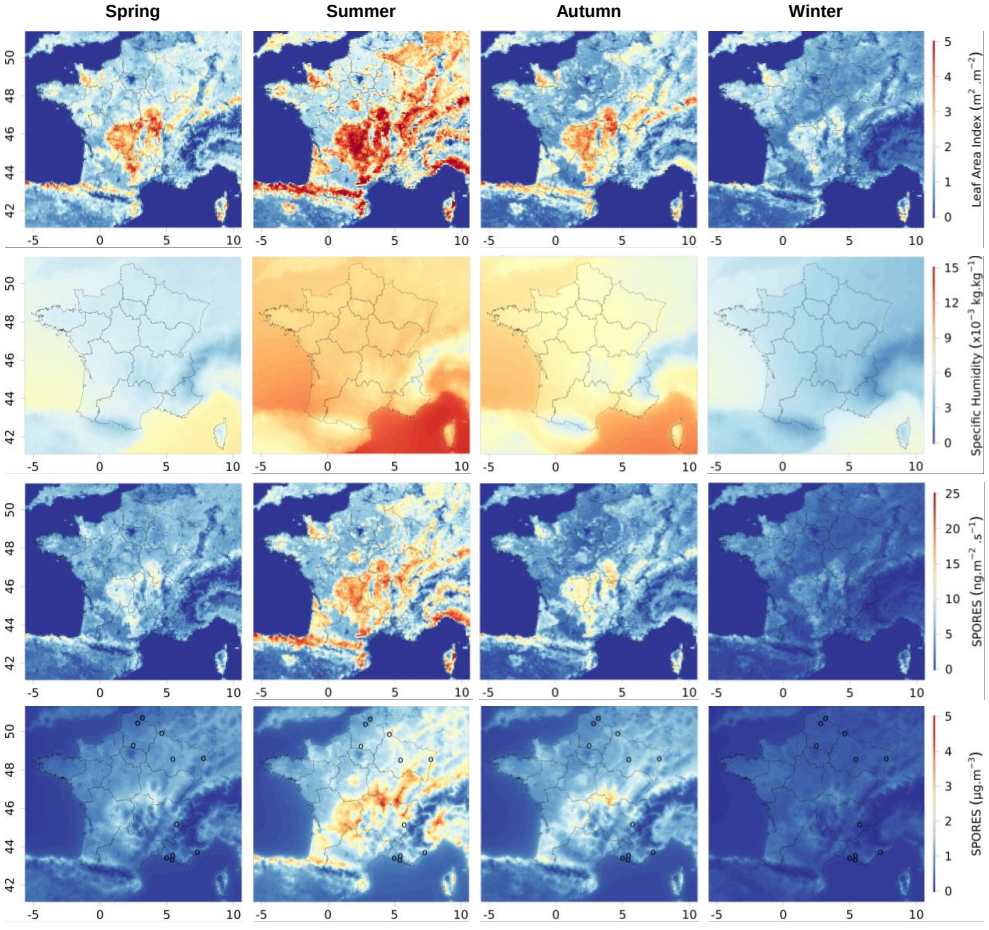

Figure 2 : Leaf Area Index (LAI), specific humidity, mean seasonal emissions and seasonal concentrations of fungal spores modelled with CHIMERE for 2013 and 2014 in France respectively from top to bottom, by season for spring (March to May), summer (June to August), autumn (September to November), and winter (December to February), respectively from left to right. The circles represent the location of the measurement sites

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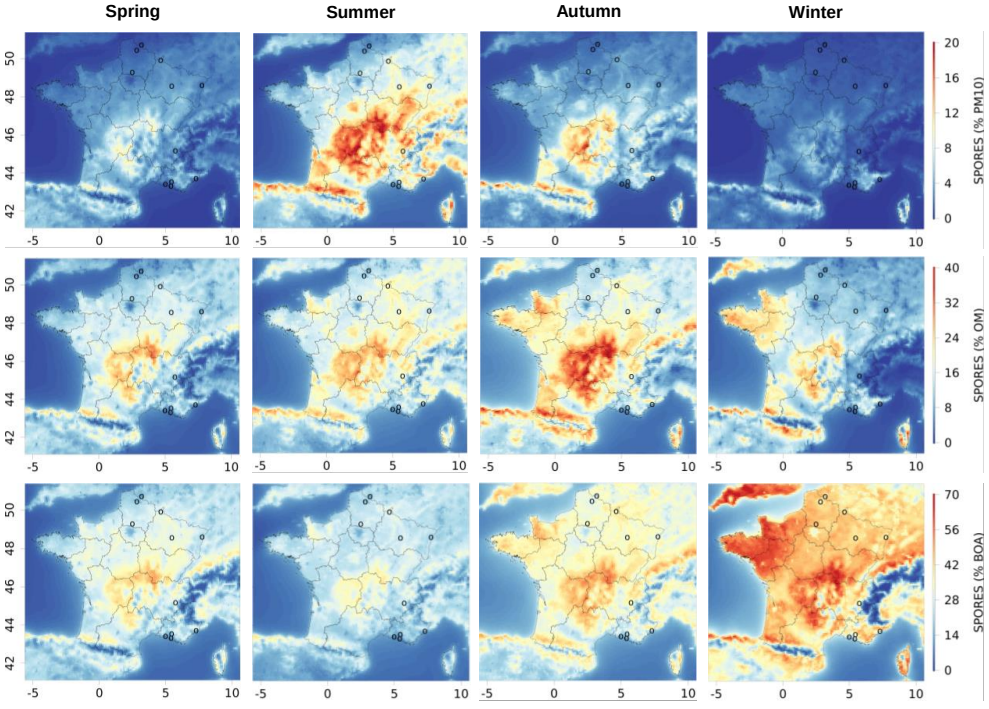

Figure 3 : Contribution of fungal spores organic matter to PM_{10}, OM and biogenic organic aerosols (BOA) modelled with CHIMERE for 2013 and 2014 in France respectively from top to bottom, by season for spring (March to May), summer (June to August), autumn (September to November), and winter (December to February), respectively from left to right. The circles represent the location of the measurement sites.

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3.2. Comparison of fungal spore simulations to observations

3.2.1. General comparison for the entire data set

In order to compare simulations with observations, we can rely on two types of datasets available for several sites (as described respectively in sections 2.1.1 and 2.1.2): first the polyol concentrations, and second the total OM concentration within the primary biogenic factor (OMpb) derived from PMF analysis of PM_{10} filter samples. For these comparisons to be meaningful, we need to convert the simulated fungal spore organic aerosol concentrations into polyol ones. Bauer et al. (2008b) derived a conversion factor for this purpose, for the temperate latitude continental urban site of Vienna. For the sum of arabitol and mannitol, which are the two sugar alcohol species measured in our data base, the latter authors found an average mass of 2.9 (2 – 4.2) pg per fungal spore. Elbert et al. (2007) assumed an average mass of a fungal spore of 65 pg. Combining these values yields to a polyol / fungal spore mass ratio of 4.5 (3.1 – 6.5), which will be used for the comparisons that follow. This is coherent with the work of Heald and Spracklen (2009), who used this same combination of values in order to derive their initial estimation of the mass of fungal spore emissions from multi-site polyol measurements.

We can first obtain a general picture of the performances of the model by studying the correlations and biases for all of sites with polyol measurements. For the 169 polyol monthly averages from 11 sites, the median mean fractional bias (MFB) is slightly negative (-11 %), but with a large range of values for individual sites ranging from -78 % to +53 % (Figure 4, Table S1)[1]. Using the lower and upper boundaries for the conversion factor between mass of spore and mass of polyols (3.1 and 6.5, respectively), the corresponding median MFB values would be -47 % and +26 %. As a conclusion, within the range of quantified uncertainties, the median MFB for monthly polyol means of -11 % is statistically close to zero. A bias calculation performed directly with the 1497 daily means shows very similar results, with a median MFB of -11 % (range for the -81 % to +49 %). This is not surprising, since the comparison of monthly means has been based only on days for which observations were available.

Next, simulated fungal spore OA is compared to OM in the primary biogenic factor (OMpb) (see section 2.1.2). Our simulations show a median MFB bias of -28 % and a range from -116 % to +22 % for different sites (Figure 4, data from 98 monthly means for 7 sites). A negative bias is expected for this comparison, since the PMF factor is likely to include OM contributions from BSOA in addition to that from fungal spores (see section 2.1.2). As a result of this bias analysis with two different types of observations (polyols, OMpb), we do not observe the presence of a systematic bias for our fungal spore OA simulations for the ensemble of French sites. This agrees with (Hummel et al., 2015), who also could not conclude on a significant bias of the H&S parameterisation.

[1] Note that following the definition of MFB (see SI), a relative difference between simulations and observations of a factor 2 (1/2) corresponds to a MFB of 67 % (-67 %). Thus in very crude manner, the simulations with sites with the largest and lowest MFB show a factor 2 difference with observations.

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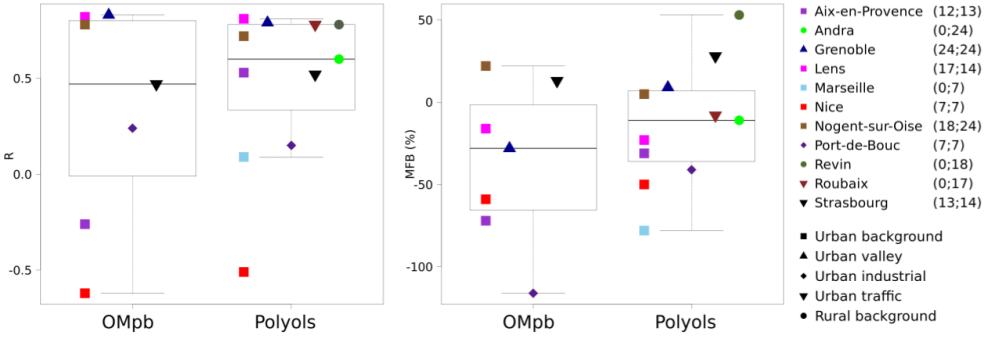

Figure 4 : Comparisons of simulated monthly mean concentrations of fungal spore OA to OM from PMF primary biogenic factor (OMpb) and polyols measurements (sum of mannitol and arabitol). Biannual mean correlation and mean fractional bias (MFB) are respectively illustrated at left and right side. Ranges between minimal and maximal values, and medians for respectively 7 and 11 sites. The number of monthly data for OMpb and polyols are noted next to the station list out of a total of respectively 98 and 169 monthly data.

The data sets were also used in order to asses if the H&S parameterisation is able to reproduce the daily and the monthly average time variation (Figure 4 and Figure S1, Table S1). For daily polyol averages, the median correlation between simulations and measurements is 0.43 with a range from -0.19 to 0.57 for the 11 sites. The median correlation is increased to 0.60 when looking at monthly averages with a large range from -0.51 to 0.83. Expectedly, for many, but not all sites, the parameterisation better depicts the seasonal variation (with larger summer and lower winter values) compared to the daily variations (Figure S1). We will discuss this result further on a site-by-site basis in section 3.2.2. 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 (Table S1, Figure S2). The root mean square error (RMSE) was also calculated for estimating the error (Table S1) the median RMSE calculated with daily polyol data is 0.04 µg m^{-3} and 0.03 µg m^{-3} with monthly data.

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3.2.2. Comparison of time series at selected sites

In this section, we evaluate the robustness of our simulations as a function of the period of the year. To do so, comparisons are conducted between model outputs and polyol observations, which are available for more measurement sites than sites with PMF results. These comparisons especially aim at understanding the large ranges of biases and correlations encountered in the previous section. Figure 5 shows observed and estimated monthly mean polyol (sum of arabitol and mannitol concentrations) at the sites with the most data (> 130 daily data) during years 2013 and 2014, namely Grenoble, Lens, Nogent-sur-Oise, Revin and Roubaix. These sites also have the advantage of being of different types, respectively urban background in an Alpine valley, urban background, urban background, rural background, and road traffic. The time series for the other sites are shown in Figure S3, Figure S4, Figure S5. We indicate both the simulated monthly means using data from all days, and only for days for which filter samples are available.

Differences between simulations and measurements are small (<10 %) for most of the values, which underlines the robustness of the model for monthly averages. Figure 5 shows simulated seasonal cycles coherent with that in Figure 2 which reflects the dependence of the simulated emissions on the LAI. We observe the maximum monthly values for the summer months with a difference in structure between 2013 and 2014: while in 2013 the simulated maximum occurs in July for all of the sites, in 2014, it occurs in September at least for the sites in Northern France (Roubaix, Lens, Nogent-sur-Oise, Andra-OPE, Strasbourg). The highest summer measurement values of polyols (0.1 – 0.15 µg m^{-3} corresponding to 2 - 3 µg m^{-3} of OMpb for monthly averages) are of course simulated on the sites where the regional LAI are the strongest (e.g. Grenoble, Andra-OPE, Revin, Strasbourg, Nogent-sur-Oise) as opposed to Lens, Roubaix, Marseille, Aix, Port-de-Bouc for which the LAI of the adjacent regions are lower. However, none of the measurement sites are located within the area of large simulated fungal spore OA concentrations over the Massif Central. Comparisons between simulations and observations show a remarkable agreement especially in the seasonal variation for the stations in the northern part of France (Lens, Roubaix, Revin, Nogent-sur-Oise), resulting in monthly correlation coefficients (R) of respectively 0.78, 0.83, 0.78 and 0.72. Specifically, the gradual increase in polyols (and related fungal spores OM) from March to July is very well simulated, except for Revin for which summer concentrations are overestimated. MFB values vary between -23 % for Lens and +53 % for Revin.

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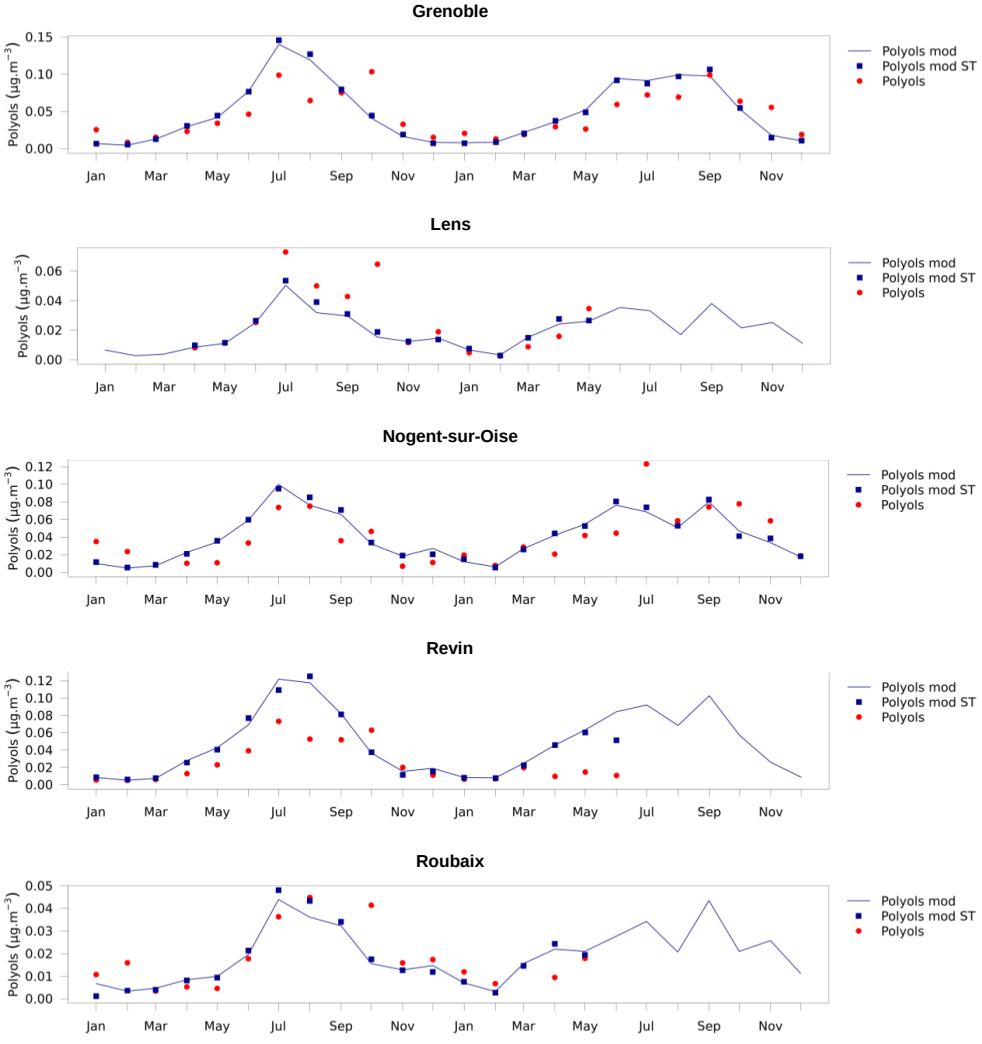

Figure 5 : Timeseries of monthly-mean polyol concentrations over 2013 and 2014 modelled by CHIMERE (blue line), measured at the sites (red dots) and modelled by CHIMERE using the same timebase as the measurements (blue squares). The simulated values by CHIMERE have been derived by using a 4.5 % conversion factor between fungal spore OA and sum of polyols. Only the sites with the highest numbers of daily data used to calculate the monthly averages are shown (Grenoble, Lens, Nogent-sur-Oise, Revin, Roubaix).

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Correlations for eastern French sites are a bit lower, with 0.60 for the Andra-OPE site and 0.52 for Strasbourg with MFB respectively of -11 % and +28 %. For Grenoble, a city in SE France within the Alps, correlation is good (R = 0.79) and bias is small (MFB = +9 %). For a group of sites in the south of France (Port-de-Bouc, Marseille, Nice), located less than 10 km from the Mediterranean Sea, the situation is singularly different, with strong underestimations in the simulation. It should also be noted that we have fewer observations at these sites (only seven monthly mean observations from June to December 2014), meaning that a full seasonal cycle was not obtained. Still, the simulated decline in autumn/winter (October to December) compared with summer (June to August) is not observed at these sites, resulting in low or even negative correlations for monthly means between -0.51 and 0.15 and negative biases (MFB values between -41 % and -78 %). Similarly, for Aix-en-Provence, some 30 km inland, winter polyol levels are strongly underestimated, resulting in a MFB of -31 % and a correlation of 0.53.

3.3. Discussions

Overall, results obtained in this study demonstrate that the H&S parameterisation implemented into the CHIMERE model works remarkably well to reproduce the concentrations of fungal spore OA (or at least a proxy of these concentrations, with the polyols measurements) observed at sites located in the northern (Lens, Roubaix, Revin, Nogent-sur-Oise) and eastern (Andra-OPE, Strasbourg, Grenoble) parts of France. Indeed, the seasonal cycles observed at these sites and the intensity of the concentrations are remarkably well simulated by the model for the monthly averages. This gives great confidence in the ability of the H&S parameterisation to reproduce the fungal spore OA source over large parts of France. This extends the results from the earlier work of Hummel et al. (2015) based on an evaluation of 4 sites located in more northerly parts of Europe (Finland, Ireland, UK, Germany) limited to a week in the end of August, to more southerly regions, but still with temperate vegetation, and full seasonal cycles. For Europe, this extends also the results from Janssen et al. (2021) who implemented the H&S parameterisation into the global GEOS-Chem model. They compared the model output to yearly FBAP observations at the same sites in Finland and Germany and found rather similar seasonal variations with summer maxima and winter minima, although the simulated maximum occurred in June (2010), while it was observed in August. Note that Janssen et al. (2021) shows that the H&S parameterisation shows a strong overestimation of fungal spore numbers with respect to observations in the US.

Another remarkable fact is that positive results in our study have been obtained from sites with very different land-use typologies, ranging from traffic (Roubaix and Strasbourg) and urban background (Lens, Nogent-sur-Oise) to rural (Revin, Andra-OPE), or an urban background site within an Alpine valley (Grenoble). This can be explained by the fact that, due to low levels of long-distance transport, fungal spore OA seems to be controlled by the vegetation at local scale, as also pointed out already for Grenoble by Samaké et al. (2019a).

Despite these overall encouraging results, several limitations appear for our study. One is probably related to the simplification of using a unique LAI parameter

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which cannot consider differences in vegetation typology. This may explain strong differences in MFB values between sites in NE France: Revin, located in a forest rich area in the Ardennes, shows a strong positive MFB of +53 % (the largest one encountered in our study), while the Andra-OPE site surrounded by extensive field crops shows an MFB of -11 %. For this latter site, we also can note that several observed daily peaks (in August 2013 and July 2014), as large as 5 µg m^{-3} are not simulated. Such peaks may be related to agricultural activities such as harvesting as demonstrated by Samaké et al. (2019b) from the record of field work. In addition, atmospheric concentrations of fungal spores mainly come from plant host species (Samaké et al., 2020), so mechanised crop pruning and harvesting can have an impact on spore concentrations in rural areas. The processes which are known to trigger fungal spore emissions are not included specifically in the H&S parameterisation. In the context of this work, we did not seek to better characterise this potential missing source, but it is an interesting perspective for future work.

Our study also clearly shows the inability of the H&S parameterisation to correctly reproduce OMpb and polyol measurements for Mediterranean areas in Southern France, even though as noted before, our observational data base is weaker for this region. However, at these sites, analysis of the chemical composition of aerosols in the PM_{10} fraction also showed poor simulation of the chemical species, suggesting a more global problem in the Mediterranean area. This could be explained by the specific dynamics in this sector (sea breezes, strong mistral-type winds) coupled with significant orography and heavy urbanisation. As a result, failure to take account of wind speed in the parameterisation of H&S may be a major cause of a lack of emission and concentration in the Mediterranean area. Again, this failure may also be related to the fact the LAI does not capture specific characteristics of Mediterranean type vegetation, and which are not included in the H&S parameterisation, mainly tested for sites mostly in northern Europe. In addition, it is striking that our simulations on Mediterranean sites, as expected still simulate weak autumn/winter emissions due to low LAI and specific humidity, but which are in contradiction to the still large observed concentrations. This could be due to a relatively stronger importance of soil related fungal spore emissions, which would be independent of LAI. Further, the drier and hotter Mediterranean climate could lead to relatively smaller emissions during dry summers and relatively larger emissions during winter still warm enough to allow for fungal spore emissions. It was observed by Samaké et al. (2019b) that the sudden and large decrease of the fall concentrations to the winter levels observed simultaneously in Grenoble and Chamonix (160 km apart) coincides with a first night temperature below +5 °C, which may be a threshold for the fungi population in this area. Such complex relationships would not be captured by the single specific humidity parameter which agglomerates information from relative humidity and temperature.

Finally, it may be noted that marine sources could also contribute to enhanced polyol levels and organic aerosol at near coastal sites, although such sources are not considered in our simulation. For instance, Fu et al. (2013) reports that large mannitol concentrations, up to more than 50 ng m^{-3} over the Arctic Ocean, are comparable to the maximum concentrations observed at our Mediterranean coastal sites. They attribute this source to long range transport of fungal spores, despite the small transport distance at least in the boundary layer due to efficient dry deposition. Direct marine sources for polyols are an alternative explanation (algae, marine fungi). Particularly, mannitol can account up to 20-30 % of the dry weight of some algae

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species and is likely to be an important source of carbon for marine heterotrophic bacteria (Groisillier et al., 2015). As a conclusion, the H&S parameterisation should not be applied for PBOA emissions in Mediterranean or marine areas, and further work is needed to better document PBOA concentrations and emission processes in such areas.

4. Conclusions

In this work, we introduced the parameterisation proposed by Heald and Spracklen (2009) for fungal spore OA emissions and updated by Hoose et al. (2010) into the CHIMERE regional chemistry-transport model (Hereafter called H&S). The rationale for this work is to recognise the potentially important contribution of fungal spore to summertime PM_{10} (Samaké et al., 2019b, a) that can fill in the missing part of the OM in chemistry transport models. The simplicity of the H&S parameterisation gives us specific advantages: a unique LAI parameter gives a slow varying emission potential, which is modulated with respect to meteorological conditions by specific humidity.

Here, we largely extend the geographical and temporal validity of this parameterisation, which has only been tested before for a limited dataset of observations at northern European locations during the end of summer, to a two-year dataset of seven sites over north-eastern France. Both polyols (more precisely sum of arabitol and mannitol observations), and a primary biogenic organic aerosol factor from PMF analysis show only limited biases for these sites, respectively +5 % and -2 %, in terms of MFB (from 4 sites only for the comparison with PMF analysis). These small biases, largely within the incertitude of the polyol/OM conversion factor and of the PMF factor, are a positive outcome of our study. In addition, for this group of sites, the seasonal variation of fungal spore emissions, displaying large summer and small winter values, is correctly depicted, as manifested in large monthly mean correlations (median 0.78, range from 0.52 to 0.83, from polyol measurements).

Still, and obviously, limitations can be noted, such as a wide range of biases for individual sites, with MFB values between -23 % and +53 % for polyol observations. This might be related to biome specific differences in the emissions only described by a single LAI parameter. The emission variability on a day-to-day basis is only partly expressed by the single specific humidity parameter (range of correlation coefficients between 0.31 and 0.57 for the polyol measurements at the 7 sites in North-eastern France). Here, using a more sophisticated combination of meteorological parameters would be desirable to improve the modelling, as for example in Janssen et al. (2021) including also maximum and minimum daily temperatures and friction velocity (even if these authors did not evaluate the capacity of such a combination to simulate the daily PBOA variation). One possible reason for the lack of correlation in daily time series is the impact of land-use dependent activities, such as annual harvest or tilling in agricultural areas.

For a smaller group of Mediterranean sites, with less observational data coverage, the H&S parameterisation failed to capture fungal spore emissions both in terms of absolute values and in seasonal variations, leading to strong negative biases especially during the autumn/winter seasons. As a conclusion, for this region the use of the H&S parameterisation in regional PM modelling may not consider certain factors

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necessary for these specific sites. Additional efforts are required to enhance the model dynamics specifically over Mediterranean coastal environment. This includes extending the simulation of fungal spores over more extended periods in these locations. Furthermore, there is a need to better characterise a source of Mediterranean marine organic aerosol (AO) that is distinct from fungal spores but shares the emission of polyols. It is also necessary to have more measurement points in this specific area to be able to achieve a more concrete conclusion.

These two year-round CHIMERE simulations incorporating the H&S parameterisation revealed a significant contribution of fungal spore OA to PM_{10} mass, which is of the order of one percent or less during winter, and up to 20 % during summer in high emission zones over forested areas such as the Massif Central. In terms of contribution to OM, the simulated autumn fungal spore contribution is even as high as 40 %. This large predicted fungal spore OA contribution over the Massif Central however still warrants confirmation by observations.

Finally, the projected impact of fungal spore organic aerosol (OA) suggests significant and seasonally variable contributions to both PM_{10} and OA mass. Consequently, the simulation of spores should be included in state-of-the-art chemistry transport models. While the validity of the H&S parameterisation has been demonstrated with a good agreement with measurements across northern and eastern France, its application is cautioned against in Mediterranean regions.

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Code and data availability

All measurement and PMF data for this paper are archived at the IGE, and are available on request from the corresponding authors (JLJ and GU). The codes and modelling data are available from the LISA authors (MV, GF, MB, GS).
The model is available here: https://www.lmd.polytechnique.fr/chimere/
The MODIS observations are available here : https://modis.gsfc.nasa.gov/data/dataprod/mod15.ph

Author contributions

JLJ and GU provided the PM_{10}, polyol and PMF speciation data developed at the IGE for the PhD work of Abdoulaye Samaké and Samuel Weber. OF completed the data set with those obtained at the LCSQA during the CARA programme. FC developed the H&S parameterisation code at INERIS, GS adapted the code for a more recent version of the CHIMERE model at LISA. AC contributed to the LAI mapping. MV, GF, MB, designed the numerical experiments. MV performed the simulations, produced figures and tables, and wrote the paper. All co-authors contributed to the discussion of the results. MV prepared the paper with contributions from all co-authors. MV, MB, GF, GS, JLJ and GU designed the study. MV, MB, GF, GS, JLJ, GU, OF, FC and AC contributed to the writing of the article.

Competing interests

The authors declare that they have no conflict of interest.

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