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Implementing marine organic aerosols into the GEOS-Chem model

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Abstract

Marine organic aerosols (MOA) have been shown to play an important role in tropospheric chemistry by impacting surface mass, cloud condensation nuclei, and ice nuclei concentrations over remote marine and coastal regions. In this work, an online marine
primary organic aerosol emission parameterization, designed to be used for both global and regional models, was implemented into the GEOS-Chem model. The implemented emission scheme improved the large underprediction of organic aerosol concentrations in clean marine regions (normalized mean bias decreases from −79 % when using the default settings to −12 % when marine organic aerosols are added). Model predictions
were also in good agreement (correlation coefficient of 0.62 and normalized mean bias of −36 %) with hourly surface concentrations of MOA observed during the summertime at an inland site near Paris, France. Our study shows that MOA have weaker coastal-to-inland concentration gradients than sea-salt aerosols, leading to several inland European cities having > 10 % of their surface submicron organic aerosol mass concentration with a marine source. The addition of MOA tracers to GEOS-Chem enabled us to identify the regions with large contributions of freshly-emitted or aged aerosol having distinct physicochemical properties, potentially indicating optimal locations for future field studies.

1   Introduction

With the decrease in anthropogenic emissions of particulate matter in many industrialized countries, an increased emphasis has been placed on understanding the inventory of natural aerosol sources (Zare et al., 2014). Natural aerosols also have an important climatic impact, as long term changes in emissions and the feedbacks on meteorology can lead to a global mean radiative perturbation approaching $1\,\mathrm{W\,m^{-2}}$ (Carslaw
et al., 2010). Marine organic aerosols (MOA), which have been observed at concentrations $> 1.0\,\mathrm{\mu g\,m^{-3}}$ (Ovadnevaite et al., 2011) and whose estimated global emissions

are comparable to that of fossil fuel burning (Spracklen et al., 2008), are one type of natural aerosol with air quality and climate significance (Gantt and Meskhidze, 2013). For instance, MOA have been shown to affect the surface mass, cloud condensation nuclei, and ice nuclei concentrations in clean marine regions (O'Dowd et al., 2004; Meskhidze et al., 2011; Westervelt et al., 2012; Burrows et al., 2013; Partanen et al., 2014). Uncertainty in the chemical composition of sea spray aerosol (SSA) has also been shown to play an important role in determining their climate impact (Tsigaridis et al., 2013). Therefore, it is important for chemical transport and climate models to take MOA emissions and physicochemical processes into consideration.

With instrumentation such as the high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS), source profiles of ambient organic aerosols can be derived using positive matrix factorization (PMF) techniques (Lanz et al., 2007; Zhang et al., 2011). Chang et al. (2011) derived a marine biogenic PMF factor based on high levels of organic sulfur species observed at several coastal locations affected by marine air masses. Applying HR-ToF-AMS/PMF analysis similar to Chang et al. (2011) to chemically-identify MOA, Crippa et al. (2013a) found that on average 16 % of the total summertime organic aerosol mass ∼ 20 km SW of Paris had a marine source despite being > 150 km from the English Channel and Atlantic Ocean. Concurrent summertime HR-ToF-AMS measurements in the core of the Paris metropolitan area reported by Crippa et al. (2013b) indicated that 13 % of the total OA had a marine source. Other studies have identified similar MOA-specific HR-ToF-AMS mass spectra in other locations (Ovadnevaite et al., 2011; Schmale et al., 2013) to better understand its sources. Unlike typical organic aerosol observations, these HR-ToF-AMS spectra allow for the model evaluation of marine organic aerosol concentrations separate from the surrounding terrestrial/anthropogenic emissions. Although some mathematical mixing between sources cannot be excluded in the HR-ToF-AMS/PMF analysis, Crippa et al. (2013b) obtained similar MOA mass concentrations and percentage contributions when HR-ToF-AMS measurements were combined with that of a high sensitivity proton transfer reaction mass spectrometer (HS-PTR-MS).

Although secondary organic aerosol (SOA) precursors can have a marine source (Shaw et al., 2010), marine primary organic aerosols (POA) have been shown to be the major contributor to organic aerosol mass concentrations over marine regions (Gantt and Meskhidze, 2013). The observation of clean marine organic aerosol surface con-
5 centrations having the same seasonal cycle as that of surface chlorophyll *a* concentrations ([chl *a*]) (Cavalli et al., 2004; Sciare et al., 2009) led to the development of marine POA emission parameterizations based on a [chl *a*]-derived organic mass fraction of SSA (OM_{SSA}) (O'Dowd et al., 2008; Vignati et al., 2010). Gantt et al. (2012) compared several marine POA emission schemes in a global model, finding that the schemes
10 with a strong dependence on [chl *a*] had concentrations most similar to seasonal observations. However, the use of [chl *a*] as a proxy for OM_{SSA} has come into question by several laboratory- and field-based studies showing that oceanic organic carbon concentration might be more closely related to organic enrichment of SSA (Prather et al., 2013; Quinn et al., 2014). Burrows et al. (2014) modelled the organic enrichment of
15 SSA using several classes of organic compounds from a biogeochemical ocean model, finding that OM_{SSA} is related to [chl *a*] only in certain regions. Rinaldi et al. (2013) found that [chl *a*] was more highly correlated with OM_{SSA} than oceanic organic carbon, but suggested an 8-day time lag to account for biological processes responsible for the production of transferable organic materials during the phytoplankton bloom evolution.
20 Recent field studies have suggested that other physical/biological processes affecting sea spray aerosol production may also be missing in current emission parameterizations, such as the diurnal variability in sea spray aerosol generated from biologically productive waters (Long et al., 2014). Like most global estimates of marine POA emissions, we use satellite-derived [chl *a*] as a proxy for OM_{SSA} due to the lack of consensus
25 of factors driving organic enrichment of sea spray aerosol, strong correlations observed between organic aerosol concentrations and [chl *a*] in clean marine environments, and scarcity of global oceanic datasets for use in chemical transport models.

In addition to emissions, the physicochemical treatment of terrestrial and marine organic aerosols in the atmosphere affects their predicted concentrations. A summary of

past modeling studies showed that the aging of marine and terrestrial organic aerosols (calculated either by e-folding times, microphysics, or oxidant/sulfate concentrations) is highly variable spatially and temporally with global averages ranging from ∼ 1 to 5 days (Huang et al., 2013). Spracklen et al. (2008) found that doubling the marine organic aerosol e-folding time for conversion from hydrophobic to hydrophilic in GEOS-Chem (thus decreasing the aerosol aging process) from 1.2 to 2.4 days increased surface MOA concentrations by ∼ 15 % globally due to decreased in-cloud scavenging. Westervelt et al. (2012), on the other hand, did not predict significant differences in surface concentrations after changing the aging timescale of marine organic aerosols from 1.5 to 4.5 days in the GISS II-prime model. Huang et al. (2013) found that several detailed aging schemes (including oxidant and condensation-coagulation aging) for terrestrial carbonaceous aerosols in GEOS-Chem led to a variable hydrophobic to hydrophilic conversion lifetime, ranging from < 1 to 8+ days. Modeling studies have typically treated the chemistry and physics of marine organic aerosols within the atmosphere like that of terrestrial organic aerosols due to the lack of understanding of marine boundary layer aerosol processes (Meskhidze et al., 2013).

In this study, we use the GEOS-Chem model to quantify the contribution of marine POA emissions to the global surface organic aerosol mass concentration. GEOS-Chem is a global chemical transport model widely used for simulating aerosol and gas emission processes, atmospheric chemistry, regional and global scale pollution transport, and for providing boundary conditions to regional chemical transport models (CTMs). Global emissions estimates of marine POA and inter-comparison of multiple emission parameterizations has been previously performed using older versions of GEOS-Chem (Spracklen et al., 2008; Lapina et al., 2011; Gantt et al., 2012); these studies had variable success replicating the observed surface organic aerosol concentrations in clean marine environments. Nevertheless, analysis of model results indicates that addition of marine primary organic source brought model results closer to observations. Despite these previous studies, the current version of the GEOS-Chem model does not include online marine POA emissions in the default settings. The overall objective of this study

is to implement an online marine POA emission parameterization into the current version of GEOS-Chem (v9-02) that can be easily used in the default setting with the following characteristics: (1) adds minimal computational expense, (2) capable of being used for all GEOS-Chem model domains/simulation periods, and (3) treated with unique tracers capable of explicit atmospheric aging and tracking. During this study the emission parameterization is tested for the global and nested regional model domains and evaluated with new datasets having advanced organic chemical characterization and widespread global coverage. Finally, the model is used to predict global surface concentrations, ocean-land concentration gradients, and relative contributions of nascent (freshly emitted) and aged marine organic aerosols.

2   Model configuration

2.1   GEOS-Chem

We use v9-02 of the global CTM GEOS-Chem (http://geos-chem.org/) with $2° \times 2.5°$ (latitude–longitude) horizontal resolution and 47 vertical hybrid sigma-pressure levels, driven by Goddard Earth Observing System (GEOS)-5 assimilated meteorology from the NASA Global Modeling Assimilation Office (GMAO). The model is run with a full chemistry configuration, which includes H_2SO_4-HNO_3-NH_3 aerosol thermodynamics (ISORROPIA II) coupled to an O_3-NO_x-hydrocarbon-aerosol chemical mechanism (Bey et al., 2001; Park et al., 2004; Fountoukis and Nenes, 2007). Terrestrial emissions of carbonaceous aerosols are based on Bond et al. (2007) (biofuel and fossil fuel) and daily wildfire emissions from version 3 of the Global Fire Data (GFED3) database (van der Werf et al., 2010). Secondary organic aerosol formation is included in our simulations based on Pye et al. (2010). Although marine-source SOA have been shown to contribute to the organic aerosol mass concentration in some regions (Decesari et al., 2011; Fu et al., 2013; Hu et al., 2013), we do not consider oceanic emissions of SOA precursors because they have uncertain emission inventories (Sinreich et al., 2010;

Miyazaki et al., 2014), SOA formation mechanisms that are not well known (Bikkina et al., 2014), and have been estimated to contribute less than primary sources to the organic aerosol budget in many marine regions (Arnold et al., 2009; Fu et al., 2013). Sea-salt aerosols in the model are emitted in two size bins (fine mode ranging from 0.02 to 1.0 µm in diameter and coarse mode ranging from 1.0 to 16.0 µm in diameter) as a function of a power relationship with 10 m winds speeds (U_{10}) following the formulation of Gong (2003) and includes the 3rd order polynomial dependence on sea surface temperature (SST) as described by Jaeglé et al. (2011). Within this model setup, we introduce a hydrophilic and hydrophobic tracer for marine POA. The differences between the hydrophilic and hydrophobic tracers involve depositional processes: scavenging in convective updrafts and rainout only occurs for hydrophilic tracers (Liu et al., 2001). Dry deposition in the model is based on the resistance-in-series scheme described in Wesely (1989), with the surface resistances for aerosols following the work of Zhang et al. (2001). A detailed description of the various marine POA emission schemes and model treatment of the marine POA tracers is given in Sect. 2.2. A year-long GEOS-Chem simulation for 2009 is performed for the global domain, with a nested simulation of the European domain (0.5° × 0.67°) performed for July 2009 in order to show ocean-continental concentration gradients and compare with novel measurements of marine organic aerosol collected near Paris, France. 3 h dynamic boundary conditions for the nested model run are prescribed from the global GEOS-Chem simulation.

2.2 Marine POA emission

Submicron marine POA emissions implemented into GEOS-Chem are based on the top-down parameterization developed by Gantt et al. (2012), which compared several marine POA emission schemes using an older version of GEOS-Chem (v8-01-01). The top-down parameterization from Gantt et al. (2012) updated the Gantt et al. (2011) emission scheme by: (1) increasing the OM_{SSA} dependence on [chl a] and U_{10} to strengthen the correlation between model-predicted and observed organic aerosol surface concentrations at Mace Head, Ireland and Amsterdam Island over several years,

and (2) scaling the total marine POA mass emission rate (E_{POA}) to minimize the bias of GEOS-Chem-predicted surface concentrations with seasonal observations at Mace Head and Amsterdam Island. In addition to improving the prediction of seasonal observations, we use the Gantt et al. (2012) top-down parameterization because it yielded better predictions of both monthly and seasonal concentrations of organic aerosol over coastal regions, and hourly surface concentrations during a MOA plume event (Ovadnevaite et al., 2011). The Gantt et al. (2012) top-down emission parameterization was given as follows:

$$
\begin{aligned}
OM_{SSA}(chl\ a, U_{10}, D_p) = \frac{\left(\frac{1}{1+\exp(3(-2.63[chl\ a])+3(0.18(U_{10})))}\right)}{1+0.03\exp(6.81D_p)} \\
+ \frac{0.03}{1+\exp(3(-2.63[chl\ a])+3(0.18(U_{10})))}
\end{aligned} \tag{1}
$$

$$
E_{POA}(chl\ a\ U_{10}D_p) = 6 \times V_{SSA} \times OM_{SSA} \times \rho_{SSA}, \tag{2}
$$

where D_p is the sea spray particle dry diameter (µm), V_{SSA} is the volume emissions ($cm^3\,m^{-2}\,s^{-1}$) of sea spray aerosol according to the Gong et al. (2003) source function with SST dependence of Jaeglé et al. (2011), ρ_{SSA} is the apparent density ($g\,cm^{-3}$) of the sea spray aerosol calculated as a function of the organic and sea-salt mass fractions, E_{POA} has units of molecules $cm^{-2}\,s^{-1}$ after conversion from $g\,m^{-2}\,s^{-1}$ using a molecular weight of carbon, and [chl a] and U_{10} have units of $mg\,m^{-3}$ and $m\,s^{-1}$, respectively.

The daily-average [chl a] used in this study to calculate marine POA emissions in GEOS-Chem is from temporally-interpolated, monthly-average MODIS/Aqua-derived [chl a] at 1/12° horizontal resolution that is spatially-averaged online to fit the global and European domains. Although the year 2009 was simulated for this study due to the availability of marine organic aerosol measurements (Crippa et al., 2013a), model-ready MODIS/Aqua [chl a] inputs were generated for 2005–2011 and can easily be

expanded to include additional years/satellite datasets. Similar to terrestrial primary organic aerosols in GEOS-Chem, marine POA are emitted as hydrophobic and converted to hydrophilic in the atmosphere with an e-folding time of 1.15 days (Cooke et al., 1999). This is consistent with the observation that freshly-emitted submicron ma-

⁵ rine primary organic aerosols are water insoluble colloids and aggregates (Facchini et al., 2008; Collins et al., 2013) but can become more water soluble through atmospheric aging (Rinaldi et al., 2010; Decesari et al., 2011). Marine POA is emitted as an external mixture with sea-salt aerosols based on evidence that organics exist separately from sea-salt in aerosols below 200 nm in diameter (Bigg and Leck, 2008; Prather et al.,

¹⁰ 2013), although the implementation of MOA tracers allows for future changes in the chemical treatment.

3 Results

3.1 Seasonal concentrations

Figure 1 shows surface averaged (~ 100 m above ground level) MOA mass concen-
¹⁵ trations (left column) and the contribution of marine-source organic aerosol to total (terrestrial + marine) submicron organic aerosol surface mass (right column) predicted by GEOS-Chem. Simulations show that vast regions of the Northern Atlantic, Northern Pacific, and Southern Oceans have summertime concentrations (up to 1000 ng m^{-3}) up to a factor a 5 higher than the wintertime concentrations (< 200 ng m^{-3}) due pri-
²⁰ marily to the increase in emissions (see Fig. S1 in the Supplement) associated with the seasonal cycle of [chl *a*]. This strong seasonal cycle of organic aerosol concentrations is consistent with long-term observations at several mid-latitude coastal locations (Cavalli et al., 2004; Spracklen et al., 2008; Sciare et al., 2009) having summertime organic aerosol concentrations a factor of 2 to 5 higher compared to wintertime. Simi-
²⁵ larly, the MOA factor observed in Paris in the summertime by Crippa et al. (2013a, b) was not detected in the wintertime (Crippa et al., 2013b, c) because the contribution to

total OA was below the detection limit of $\sim 20\,\text{ng}\,\text{m}^{-3}$ (DeCarlo et al., 2006). According to Fig. 1, equatorial oceanic regions reveal little seasonal variation in marine organic aerosol concentrations, with low concentrations over oligotrophic oceans and high concentrations over productive coastal regions throughout the year. Inland regions far from the ocean have very low MOA concentrations ($< 50\,\text{ng}\,\text{m}^{-3}$) throughout the year, while coastal areas typically have concentrations up to $200\,\text{ng}\,\text{m}^{-3}$.

Figure 1 (right column) shows that marine-source organic aerosols typically contribute $> 80\%$ of the total (terrestrial + marine) submicron organic aerosol burden over remote oceanic regions such as the Southern Ocean and Equatorial Pacific for much of the year. In coastal regions downwind of terrestrial aerosol sources, the ratio of marine-source to total submicron organic aerosols (F_{MOA}) is much lower ($< 40\%$) than remote marine regions. Figure 1 also shows that the seasonal cycle of F_{MOA} for many inland and coastal regions (i.e., Arctic Ocean coastline, European continent) is different from that of surface concentrations (left column) because of the strong seasonal cycle of terrestrial organic aerosol concentrations from biomass burning and SOA formation. Compared to a previously-published map of F_{MOA} from the TM4-ECPL model (see Fig. 4c in Myriokefalitakis et al., 2010), GEOS-Chem predicts higher values (up to 20% vs. $< 5\%$) widespread over terrestrial regions. Similarly, GEOS-Chem predicts higher F_{MOA} than Myriokefalitakis et al. (2010) over most remote oceanic regions ($> 60\%$ vs. 10–50%). These discrepancies are likely caused by a number of differences including marine POA emission schemes (Gantt et al., 2012 vs. Vignati et al., 2010), SOA formation mechanisms, atmospheric aging schemes of hydrophobic POA, and the inclusion of supermicron terrestrial organic aerosols in F_{MOA} by Myriokefalitakis et al. (2010).

The nested GEOS-Chem simulation in this study highlights the MOA concentration gradient from coastal to inland regions. Figure 2a shows a sharp concentration gradient over Europe, decreasing from 1000 to $200\,\text{ng}\,\text{m}^{-3}$ within $\sim 250\,\text{km}$ of the northern Atlantic Ocean coastline. Three major cities roughly 25, 200, and 370 km from the coast, Amsterdam, Dusseldorf, and Frankfurt, have monthly-average surface concentrations of marine organic aerosol decreasing exponentially from 670 to 280 to $180\,\text{ng}\,\text{m}^{-3}$

for July 2009, respectively (see Fig. 2). For these three cities, the modelled F_{MOA} decreases from 37 to 12 to 8 %, respectively. Although Dusseldorf and Frankfort have significantly lower F_{MOA} than Amsterdam, these inland cities still have ∼ 10 % of their submicron organic aerosol mass contributed by a marine source. GEOS-Chem predicts that of the ten largest cities in Europe, three (Istanbul, London, and Madrid) have marine-source organic aerosols making up > 10 % of the total (terrestrial + marine) surface organic aerosol concentration. The coastal gradient of marine organic aerosol concentrations is not as sharp as that of sea-salt aerosol (see Fig. S2 in the Supplement) due to the poor in-cloud scavenging of hydrophobic nascent marine organic aerosol. Relatively weak concentration gradients between the ocean and land over the western coast of Ireland suggest that measurements at Mace Head, Ireland are likely to be characteristic of the open ocean (Rinaldi et al., 2009). The sharpest marine organic aerosol concentration gradients in Fig. 2a occur in mountainous regions of Norway due to the steep terrain.

3.2 Comparison with surface observations

The time series in Fig. 2b shows a comparison of hourly marine organic aerosol surface concentrations near Paris, France during July 2009 from the observations made by Crippa et al. (2013a) and GEOS-Chem predictions. High temporally- and chemically-resolved measurements from the HR-ToF-AMS allow for the evaluation of the model-predicted MOA concentrations on an hourly time scale. Figure 2b shows that with a few exceptions, GEOS-Chem is able to capture both the magnitude and temporal variability of marine organic aerosol concentrations (correlation coefficient = 0.62 and normalized mean bias = −36 %). The period of highest observed and GEOS-Chem predicted MOA concentrations (6–9 July) also had low black carbon concentrations (< 1 µg m^{-3}) and air masses originating in the North Atlantic Ocean. Back trajectories (not shown) were derived from NOAA's hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model (Draxler and Rolph, 2014). In the days immediately following this period (10–11 July), the observed MOA concentrations remain high while GEOS-Chem predicts a

rapid decrease in concentrations; this discrepancy may be due to additional MOA formation processes missing in the model such as marine emissions of SOA precursors. Doubling the hydrophobic to hydrophilic conversion timescale in GEOS-Chem from the baseline value of 1.15 to 2.3 days results in slightly improved MOA predictions near Paris (correlation coefficient = 0.64 [vs. 0.62 for 1.15 day aging] and normalized mean bias = −26 % [vs. −36 %]). It's worth noting that Crippa et al. (2013a) suggested a possible secondary origin of this HR-ToF-AMS marine factor due to the high degree of oxygenation and strong correlation to methanesulfonic acid concentrations, although resuspended oxidized primary organic material from the ocean could not be excluded as a source.

In addition to the comparison with hourly concentrations of AMS-derived marine organic aerosol at an inland site, we have also evaluated terrestrial and total organic aerosol concentrations predicted by GEOS-Chem to a recently-published compilation of surface organic aerosol concentrations in clean marine conditions (Gantt and Meskhidze, 2013). Figure 3 shows a scatterplot of the observational data compare to GEOS-Chem surface concentrations matched by the location and month of the measurement (albeit for different years). During baseline GEOS-Chem simulations, when only terrestrial organic aerosol emissions are included (black circles on Fig. 3), a strong model underprediction (normalized mean bias = −79 %) and poor correlation (0.16) was determined. Including marine organic aerosols in the comparison (red circles on Fig. 3) substantially reduced the model bias (normalized mean bias = −12 %) and slightly improved the correlation (0.28). Unlike the direct spatiotemporal comparison with the Crippa et al. (2013a) dataset, the weak model correlation with the extensive dataset of clean marine aerosol concentrations is not surprising because particular observations spanning several decades (1973 to 2009) are compared to monthly-averaged GEOS-Chem predictions for 2009.

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3.3 Marine organic aerosol aging

The atmospheric aging of organic aerosols, especially those with a marine source, is not well understood and is an active area of research. In GEOS-Chem, we model the aging of MOA based on the e-folding conversion from nascent (hydrophobic) to aged (hydrophilic) in a way similar to terrestrial primary organic aerosols. Tracking the nascent and aged fractions of marine organic aerosol concentrations provides a distribution of their physicochemical characteristics, which could help in identifying locations for future field campaigns. The percentage of aged marine organic aerosol (hydrophilic / (hydrophilic + hydrophobic) × 100) in Fig. 4 shows a strong ocean-continental gradient, with an aged fraction of 40 to 60 % over the open ocean increasing to nearly 100 % over the continents. As expected, the aged fraction is inversely related to the marine POA emission rate (Fig. S1 in the Supplement). Gradients between mostly nascent and mostly aged MOA occur over oceanic regions as well; the European region inset of Fig. 4 shows the aged fraction increasing from < 40 % in productive waters off the coast of Ireland to 80 % in oligotrophic waters off the coast of Spain.

In addition to having MOA with a range of atmospheric ages, optimal locations for future field campaigns should have concentrations greater than the detection limit of instrumentation capable of routine monitoring such as the Aerosol Chemical Speciation Monitor (200 ng m^{-3} for 30 min signal averaging; Ng et al., 2011). Figure 5 segregates areas in which marine organic aerosol mass concentration greater than 200 ng m^{-3} are found in both low- and highly-aged regimes (arbitrarily chosen as < 40 and > 60 % aged, respectively) as predicted by GEOS-Chem. Regions with high concentrations of low-aged MOA typically occur along coastal areas, while regions with high concentrations of aged MOA are located either inland (where MOA can be difficult to differentiate from terrestrial organic aerosols) or over oligotrophic oceanic regions (where few studies have been conducted). As many previously conducted field campaigns focused on the physical and chemical characteristics of marine organic aerosols have taken place

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in biologically-productive coastal areas (Cavalli et al., 2004; Decesari et al., 2011; Russell et al., 2010), nascent MOA have likely been sampled more frequently than aged aerosols. Regions identified as having a high concentration of aged MOA such as the Equatorial Atlantic Ocean and eastern Equatorial Pacific Ocean would be good candidates for field campaigns as they likely have MOA with physicochemical characteristics different than that of nascent aerosols. Figure 5 also identifies regions like the Arabian Sea and Bay of Biscay that have a seasonal cycle of low- and highly-aged regimes, making them good candidates for long-term field studies.

4   Conclusions

In this work, an online emission parameterization of submicron marine POA has been implemented into the GEOS-Chem model and evaluated with novel datasets of episodic events and global surface concentrations. This computationally-inexpensive marine POA emission scheme includes marine organic aerosol tracers that are independent from terrestrial tracers and treats their hydrophilic-hydrophobic conversion in the atmosphere. The flexibility of this implementation for multiple years/model domains allows for users to apply these emissions in the default setting of GEOS-Chem with minimal effort. The comparison with HR-ToF-AMS MOA observations shows that GEOS-Chem replicates the variability (correlation coefficient = 0.62) and magnitude (normalized mean bias = −36 %) of summertime concentrations at an inland site near Paris, France. When GEOS-Chem is compared to globally-distributed organic aerosol observations in clean marine conditions, the underprediction for the default setting of only simulating terrestrial emissions (normalized mean bias = −79 %) is reduced with the inclusion of marine POA emissions (normalized mean bias = −12 %). In addition to improving the predictions of organic aerosol surface concentrations, the inclusion of marine emissions allows for the prediction of the global distribution of nascent and aged MOA. With the increasing focus on background aerosol concentrations at remote marine, coastal, and inland sites, this emission parameterization has the flexibility and

ease of use to be considered for the default setting of global chemical transport/climate models such as GEOS-Chem.

This implementation of marine POA emissions in GEOS-Chem improves the prediction of clean marine organic aerosol concentrations, although additional drivers of marine POA emissions not considered here (such as oceanic organic carbon, sunlight, and/or organic composition) may be needed to refine the emission scheme in the future (Prather et al, 2013; Quinn et al., 2014; Long et al., 2014). The physicochemical treatment of marine organic aerosol aging identical to that of terrestrial organic aerosols also has large uncertainties, as the processes affecting aerosols in the marine boundary layer have a low level of understanding (Meskhidze et al., 2013). Marine emissions of SOA precursors may also be needed to further reduce the model underprediction of clean marine organic aerosol concentrations. Regardless of future refinements, this implementation of MOA into GEOS-Chem addresses a missing aerosol source, improves the prediction of clean marine and inland marine-sourced organic aerosol concentrations, and enables the model to indicate potential locations for future field studies focused on sampling marine organic aerosols.

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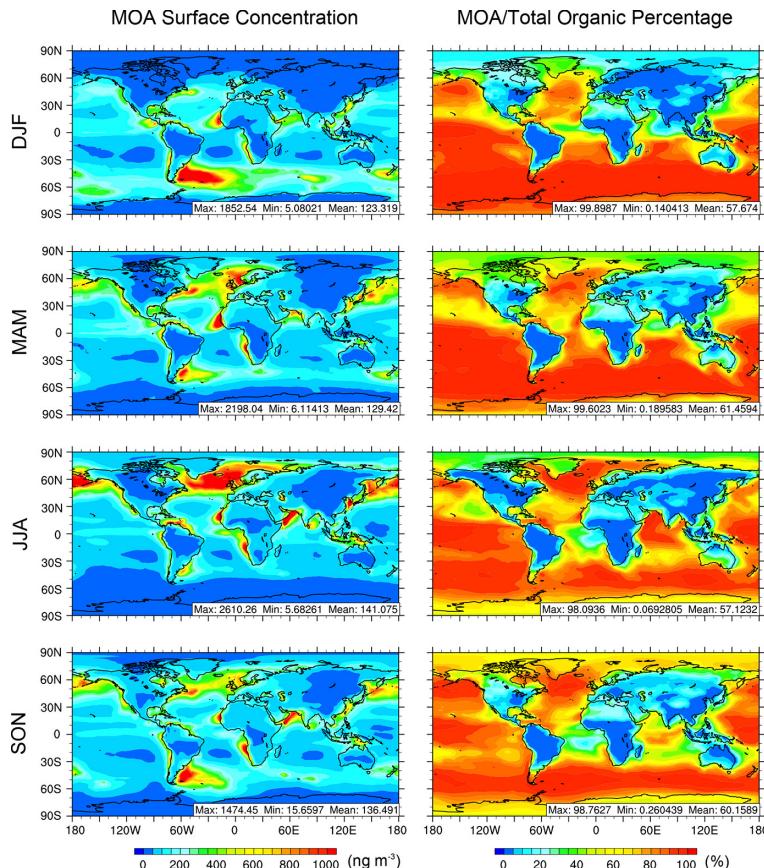
Figure 1. Seasonally-averaged submicron MOA surface concentrations and percentages of total submicron organic aerosol (marine + primary anthropogenic + biomass burning + secondary) with a marine source as predicted by GEOS-Chem for 2009.
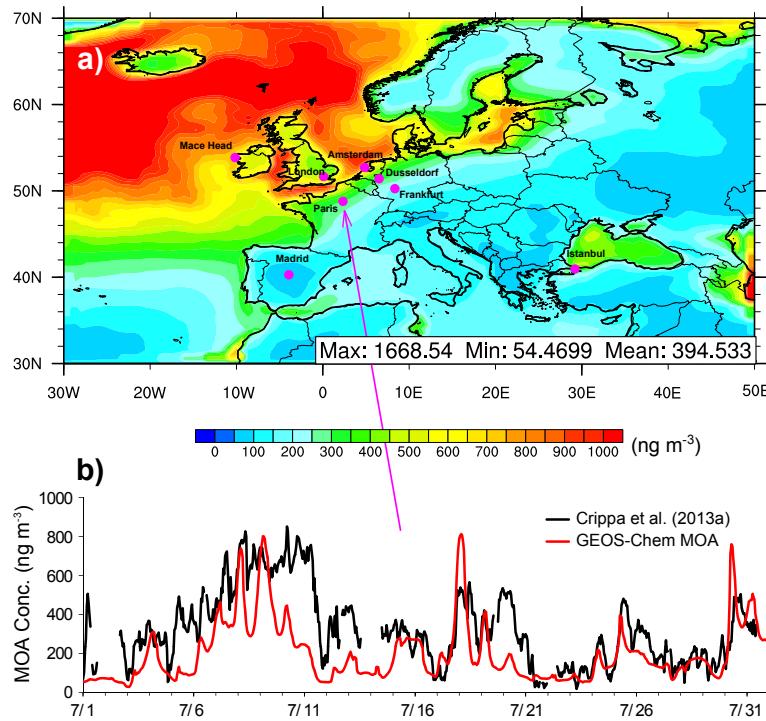
Figure 2. (a) Average submicron surface concentration of MOA for July 2009 in the nested Europe GEOS-Chem domain and the **(b)** time series of the observed and predicted marine organic aerosol concentration near Paris, France as reported by Crippa et al. (2013a).

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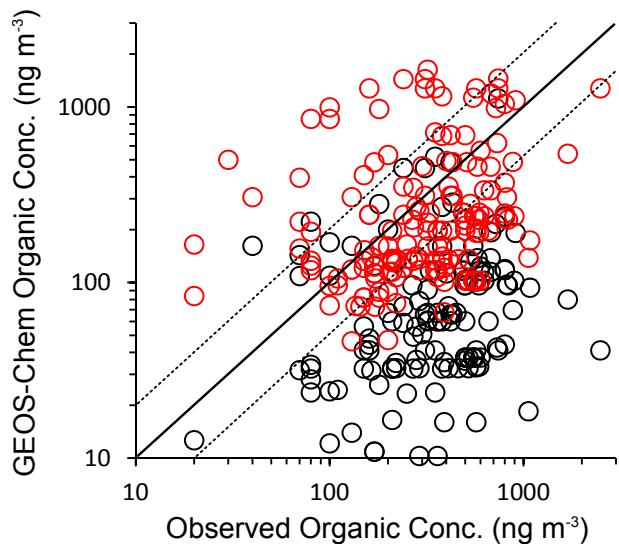
Figure 3. Global comparison of observed clean marine organic aerosol concentrations (see Gantt and Meskhidze, 2013, for details) and GEOS-Chem-predicted terrestrial (black) and total (marine + terrestrial, in red) submicron organic aerosol concentrations. Concentrations are matched by location and month(s), but represent different years (observations span 1973–2009 while GEOS-Chem predictions are for 2009). The solid and dotted lines represent the 1 : 1 line and 1 : 2 and 2 : 1 lines, respectively.

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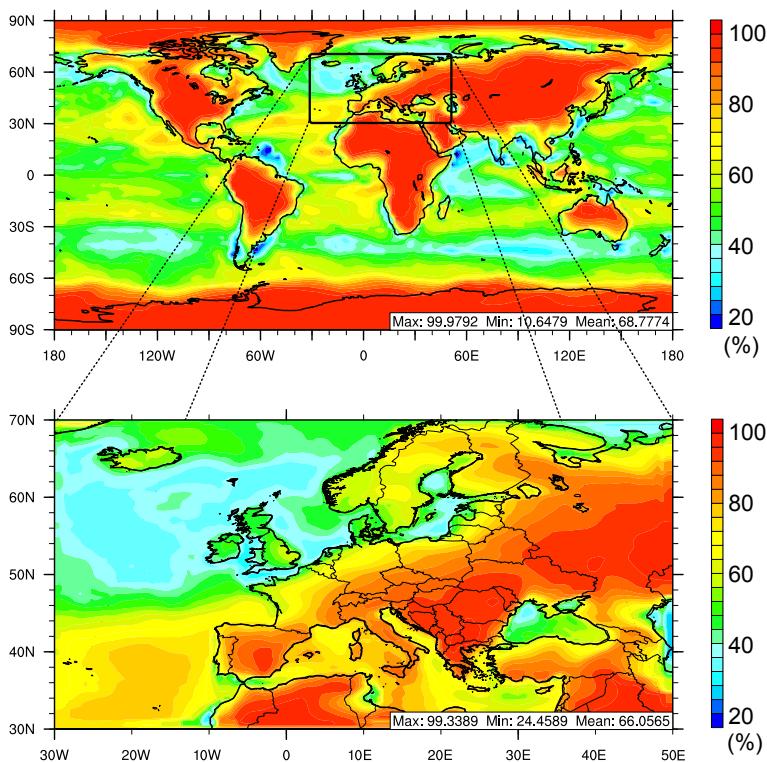
Figure 4. Percentage of the submicron MOA concentration predicted by GEOS-Chem to be hydrophilic (aged) for July 2009.

Figure 5. Regions (in red) with GEOS-Chem predicted seasonal submicron MOA concentrations $> 200\,\mathrm{ng\,m^{-3}}$ for both low-aged (left column, $< 40\,\%$ hydrophilic marine organic aerosol) and highly-aged (right column, $> 60\,\%$ hydrophilic marine organic aerosol) regimes.