Secondary Aerosol Formation and Their Modeling

Karine Sartelet

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Abstract Measurement studies using particle into liquid sampler coupled with an ion chromatograph (PILS-IC), aerosol mass spectrometer (AMS), or aerosol chemical speciation monitor (ACSM) show a large fraction of organics and sulfate in fine particles in summer over the western and eastern Mediterranean. Different formation pathways of organic aerosol (OA), from anthropogenic and biogenic precursors of continental or marine origin, are discussed and quantified, using observations and modeling. The contribution of specific formation pathways, such as autoxidation of biogenic VOC precursors, is estimated. OA properties and their seasonal variation, in particular oxidation state and related solubility, are discussed. The temperature sensitivity of secondary OA formation differs for different aerosol modeling schemes, which is important to take into account when predicting aerosol in a future warmer climate. Over the Mediterranean, sea salt is an important fraction of the coarse aerosol and interacts with the soluble organic aerosol.

Chapter reviewed by ***Didier Hauglustaine*** *(Laboratoire des Sciences du Climat et de l'Environnement (LSCE), CEA-CNRS-UVSQ, IPSL, Université Paris-Saclay, Gif-sur-Yvette, France), as part of the book Part VI Recent Progress on Chemical Processes also reviewed by* ***Andrea Pozzer*** *(Max Planck Institute for Chemistry, Mainz, Germany)*

K. Sartelet (✉)
Centre d'Enseignement et de Recherche en Environnement Atmosphérique (CEREA), École des Ponts ParisTech, EDF R&D, Marne-la-Vallée, France
e-mail: karine.sartelet@enpc.fr

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1 Introduction

The Mediterranean region is considered as one of the prominent regions that could be detrimentally impacted by climate change with an increase in temperature and in formation of secondary aerosols (Cholakian et al., 2019a). Depending on the atmospheric circulation, natural aerosols (desert dust and sea salt) are dominant compounds of PM_{10} concentrations over the western Mediterranean (Menut et al., 2015; Rea et al., 2015; Di Biagio et al., 2016; Schepanski et al., 2016; Guth et al., 2018), which are also influenced by sporadic wildfires (e.g., Pace et al., 2003; Ancellet et al., 2016). Fine particles (PM_1 and $PM_{2.5}$) are dominated by organics (compounds formed from the atmospheric transformation of organic species) and inorganics (mostly sulfate but also nitrate, ammonium, sodium, and chloride) both over the western and the eastern Mediterranean, as shown by PILS-IC, ACSM, and AMS measurements. ACSM and AMS instruments measure the real-time chemical composition and mass loading of non-refractory aerosol compounds (sulfate, nitrate, ammonium, chloride, and organic compounds) of particles with aerodynamic diameters typically between 70 and 1000 nm (PM_1). A large fraction of these inorganic and organic aerosols (OA) is secondary (Stavroulas et al., 2019), with influences from different sources (biogenic, marine, anthropogenic: traffic, residential heating, agriculture, etc.). These fine particles affect both climate and human health. They influence the direct radiation budget mostly by scattering sunlight resulting in negative direct radiative forcing. Moreover, they can act as cloud condensation nuclei and hence modify cloud microphysical properties and lifetime. Considering health effects, oxidative stress, which is induced by the generation of reactive oxygen species (ROS), is suggested as one pathway of OA toxicity. The oxidative potential may differ for the different organic precursors. The different measurement stations where campaigns were based or long-term observations performed are displayed in Fig. 1.

Fig. 1 Map of the ground-based stations involved in campaigns mentioned in this chapter

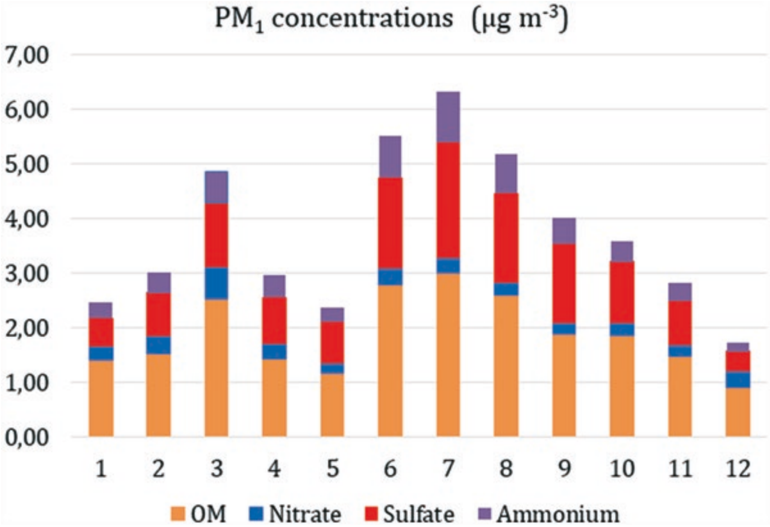

Fig. 2 Submicron aerosol composition at Ersa obtained from ACSM measurements between June 2012 and July 2014. (Data courtesy of Jean Sciare)

In summer, over the western Mediterranean, several measurement studies using ACSM or AMS stressed the high fraction of organics and sulfate in fine particles, for instance, at Montseny (Minguillón et al., 2015) and at Lampedusa (Mallet et al., 2019). Measurements performed in the framework of ChArMEx (Chemistry-Aerosol Mediterranean Experiment) at Ersa (Corsica, France) are shown in Fig. 2. Organics and sulfate represent the two main fractions of the fine aerosols observed (Arndt et al., 2017; Chrit et al., 2018a; Mallet et al., 2019), with higher concentrations during summer time. Although the fractions of organic matter (OM) and sulfate depend on air masses, during summer 2013, the PM$_1$ composition was similar between Mallorca and Ersa: organics dominate the PM$_1$ fraction (45% of OM and 40% to 42% of sulfate in PM$_1$ at both Cap Corse and Mallorca, respectively, between 10 July and 10 August 2013).

The eastern Mediterranean is also strongly influenced by sulfate and organics. Over the Aegean Archipelago, measurements performed in summer at Vigla and Finokalia showed that sulfate and OM accounted for about 40–45% and 20–32% of PM$_1$ mass, respectively (Pikridas et al., 2010; Athanasopoulou et al., 2015; Bougiatioti et al., 2016). High organic concentrations were also observed in Beirut: about 35% of PM$_{2.5}$ during the year 2014 (Abdallah et al., 2018), with higher organic fractions in summer. In July 2011, organics accounted for about 50% of PM$_{2.5}$ against about 30% for sulfate (Waked et al., 2013). Using 2-year measurements, Theodosi et al. (2018) showed that organics and inorganics make about 38% and 31%, respectively, of PM$_{2.5}$ concentrations in Athens. In summer, Stavroulas et al. (2019) measured that 46% of PM$_1$ are organics, while 31% are made of sulfate.

Secondary organic aerosols (SOA) are produced through chemical oxidation of gaseous organic compounds, among which volatile organic compounds (VOCs). The condensable products formed may then condense on organic or aqueous particulate matter, depending on their affinity with water. In summer, because solar radiation and temperature are high, a large fraction of emitted VOCs is biogenic. These biogenic VOC emissions are oxidized and are strongly affected by anthropogenic compounds, leading to the formation of SOA. The most important biogenic precursors of SOA are isoprene, monoterpenes, and sesquiterpenes. The formation of biogenic SOA is enhanced by anthropogenic plumes, because of an increase of oxidant concentrations and of absorbing mass on which SOA can condense. Therefore, reducing anthropogenic emissions may actually reduce the biogenic SOA concentration (Carlton et al., 2010; Sartelet et al., 2012). Over the United States and over Europe, Carlton et al. (2010) and Sartelet et al. (2012) estimated that as much as 50% of biogenic OA may be controllable.

In winter, the precursors of OA may predominantly be anthropogenic VOCs and I/S-VOCs (intermediate and semi-volatile organic compounds). Over Europe, the seasonality of OA due to wood burning and biogenic emissions in winter and summer, respectively, was pointed out by different studies based on either modeling (Couvidat et al., 2012) or measurements (Theodosi et al., 2018; Daellenbach et al., 2019; Yttri et al., 2019). Organic concentrations are higher during winter than summer in large cities, such as Athens (Stavroulas et al., 2019). The origins and formation pathways of these fine particles are now investigated into details using measurement and modeling studies.

2 Organic Aerosols

In order to understand the origins and formation pathways of organic aerosols, their properties are first described. For modeling purposes, it is important that not only the concentrations but also the properties of OA are well understood and represented, in order to make sure that their formation processes included in the models are as close as possible to the reality. Because of the large number of organic compounds that may partition between the gas and particle phases, they are grouped into classes in 3D air quality models. Different approaches may be used, depending on the model and/or precursor. Over the Mediterranean, two main approaches have been used to represent the compounds formed by oxidation of the precursors: a VBS approach where the classes are defined depending on the volatility of the compounds (Cholakian et al., 2018; Chrit et al., 2018b) and a surrogate approach where each class is represented by a surrogate (Chrit et al., 2017, 2018a, b). The surrogate approach differs from the VBS approach by the characterization of each class not only by the volatility of the compounds but also by the definition of specific properties (oxidation, affinity with water, molecular structure).

2.1 Organic Aerosol Properties

Carbon Origin

From experiments, the biogenic origin can be studied using high-volume quartz filter sampling ^{14}C measurements, which can determine if the organic carbon OC has a fossil or a non-fossil origin. Measurements performed over the western Mediterranean in summer suggest that most of the OA is from biogenic origins: biogenic precursors are processed and nucleate and/or condense to form OA.

At remote locations, this is illustrated by the ^{14}C measurements performed in July 2013 during the ChArMEx summer campaign at Ersa and Mallorca. OC is mainly of non-fossil origin at both Ersa (84.7 ± 3.1%) and Mallorca (70.1 ± 9.5%). We can also note the stability of the contribution of non-fossil OC at Ersa all along the summer 2013 campaign indicating a relative independence of the SOA sources toward air masses' history, while Mallorca, due to its location (low altitude and closer to anthropogenic emissions sources), is, as expected, more influenced by fossil carbon emissions, than Ersa. Even in cities, this biogenic influence dominates during summer. El Haddad et al. (2011a, b) showed, by combining data from an AMS (aerosol mass spectrometer), organic markers, and ^{14}C measurements obtained in downtown Marseille in summer, that more than 70% of the OA was from secondary and with contemporary origins. These high biogenic concentrations in an urban area are partly explained by the influence of anthropogenic emissions on biogenic SOA formation.

In modeling studies, the carbon origin is determined by tracing the SOA surrogates formed from the different precursors. Simulations performed using the surrogate and the VBS approaches over the western Mediterranean agree well with measurements performed at Ersa during summer 2013 (Chrit et al., 2017; Cholakian et al., 2018). Figure 3 shows the ratio of biogenic SOA to OA in $PM_{2.5}$ (in %) at surface during the summer 2013 using the concentrations simulated by Chrit et al. (2017). The biogenic contribution reaches 95% at remote locations and is higher than 70% over cities. The lowest biogenic SOA contribution to OA is along the ship lines (about 55%). However, at higher altitudes (300–450 m), the local anthropogenic contribution becomes lower due to slow mixing in the boundary layer over marine areas, while biogenic contributions are transported from more distant sources and are better mixed (Cholakian et al., 2018).

Oxidation

From ACSM or HR-ToF-AMS measurements, the ratios of OM/OC and O/C and the oxidation state of organics are estimated following Kroll et al. (2011). Positive matrix factorization may also be used to identify different "types" of oxidized OA (Lanz et al., 2010): HOA (hydrocarbon-like organic aerosol), LVOOA (low volatile oxygenated OA), and SVOOA (semi-volatile oxygenated OA). Positive matrix

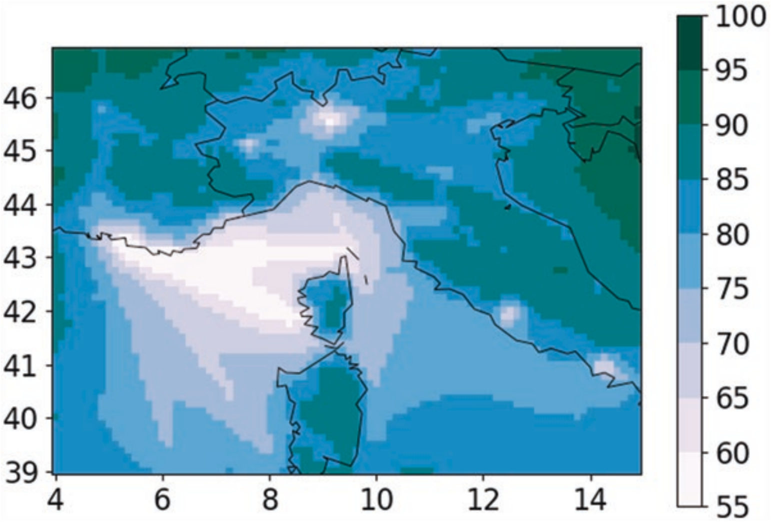

Fig. 3 BSOA/OA ratio in $PM_{2.5}$ (in %) at surface during the summer 2013, using the concentrations simulated by Chrit et al. (2017)

Table 1 Average (standard deviation) of atomic ratios derived from HR-ToF-AMS results for Ersa and Mallorca

Station	O/C	H/C	OM/OC	OSc
Ersa	0.92 (±0.11)	1.30 (±0.08)	2.34 (±0.14)	0.53 (±0.28)
Mallorca	0.64 (±0.10)	1.25 (±0.05)	1.97 (±0.13)	0.03 (±0.02)

Courtesy of Nicolas Marchand
The oxidation state (OSc) measures the degree of oxidation of organic compounds

factorization (PMF) on the combined organic-inorganic matrices separate the oxidized organic aerosol into a more oxidized organic aerosol (MOOA equivalent to LVOOA) and a less oxidized organic aerosol (LOOA equivalent to SVOOA).

In summer, at remote locations, the organic fractions of the aerosols are very oxidized (see Table 1; Chrit et al., 2017; Michoud et al., 2017; Cholakian et al., 2018; Mallet et al., 2019) with differences depending on time and location. For example, the organic fraction of the aerosols collected at Ersa in July 2013 (O:C ~ 0.92) is clearly more oxidized than at Mallorca (O:C ~ 0.64). There was a global stability of all the atomic ratios all along the campaign of July 2013 at both Ersa and Mallorca, suggesting a constant income of highly oxidized OA (LVOOA). Mallet et al. (2019) showed that at Lampedusa in June 2013, OA is also very oxidized with a ratio O:C about 1. They also found that MOOA was the dominant factor (53% of the PM_1 OA mass) and it was well correlated with sulfate.

In the framework of the ChArMEx and the ANR project SAFMED (Secondary Aerosol Formation over the Mediterranean), Freney et al. (2018) measured in late June–early July 2013 that 60–72% of PM_1 aerosols are organics and very oxidized

(O:C ~ 1.05) over forested regions in the south of France. The MOOA component was strongly associated with inorganic species, in opposition to the LOOA factor, which correlated well with biogenic volatile organic species measured with mass spectrometry, such as isoprene and its oxidation products.

To compare the observed oxidation properties to the simulations using the VBS modeling approach, Cholakian et al. (2018) distributed the different classes among HOA, LVOOA, and SVOOA: primary OA was assumed to correspond to HOA, while secondary OA was distributed among LVOOA and SVOOA depending on the saturation concentration of the class. Using the surrogate approach, Chrit et al. (2017, 2018b) and Freney et al. (2018) calculated the OM/OC and O/C ratios by weighting the OM/OC and O/C ratios of each surrogate species. Cholakian et al. (2018) found good agreement between the modeled and simulated oxidation properties after adding the formation of low volatility compounds in the VBS approach; and Chrit et al. (2017) and Freney et al. (2018) added the formation of extremely low volatility compounds (ELVOCs) from the ozonolysis of monoterpenes and organic nitrates from the monoterpene oxidation.

During winter, the oxidation state was observed to stay very high at Ersa (see Fig. 4), as well as the oxygenation level (O/C). The modeling study of Chrit et al. (2018a) managed to reproduce the high concentration observed in March 2014 at Ersa (Fig. 2) by adding emissions of intermediate and semi-volatile organic compounds (I/S-VOCs) in their model. These compounds are emitted by combustion processes (traffic and residential heating), and their volatility distribution at the emission influences the concentrations strongly. These high oxygenation levels observed in winter may stress the importance of emissions from residential heating, which may be more oxidized than those from traffic (Ciarelli et al., 2017). However, the observed organic oxidation and oxygenation states were underestimated in the

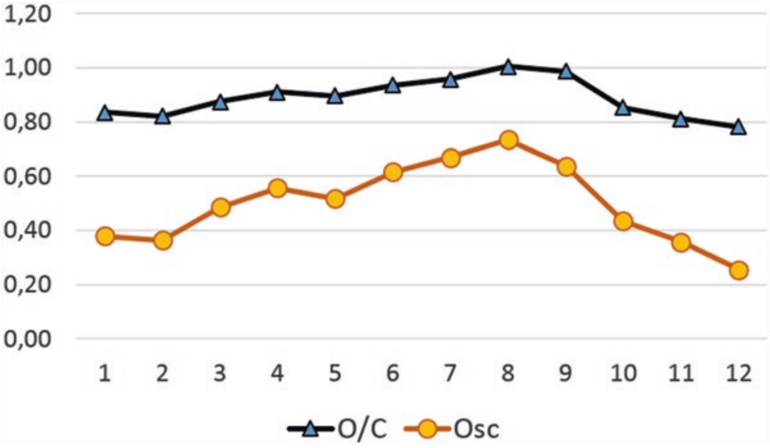

Fig. 4 Monthly averaged submicron aerosol oxidation properties (triangles, O/C ratio; circles, OSc) at Ersa (Corsica) obtained from ACSM measurements between June 2012 and July 2014. (Data courtesy of Jean Sciare)

simulations (O/C was about 0.7 in the simulations against 0.8 in the measurements), even when multigenerational aging of I/S-VOCs was modeled. This suggests that some aging processes of anthropogenic emissions still need to be elucidated, with a potential formation of ELVOCs from anthropogenic emissions (e.g., aromatics) and organic nitrate.

Solubility

Organic aerosols are observed to be highly soluble over the Mediterranean in summer. In the eastern Mediterranean, at Finokalia, Pikridas et al. (2010) estimated that at much as 80% of OC is water-soluble. At Ersa, measurements performed using a PILS-TOC-UV to estimate the water-soluble fraction of organics (Sciare et al., 2011) showed that 64% of OC is soluble (Chrit et al., 2017). Measurements using Raman lidar have also been used to characterize aerosol hygroscopicity (Navas-Guzmán et al., 2019).

Using the surrogate approach for modeling organics, the water-soluble mass of OC can be estimated from the mass of the surrogates that are water-soluble, which depends on the Henry constants of the surrogates. Chrit et al. (2017) report a good agreement between measured and simulated water-soluble organic carbon (WSOC) at Ersa (Fig. 5), with a large contribution of surrogates formed from the oxidation of biogenic precursors. The evolution of WSOC with time is due to variations in the aerosol composition depending on air mass origins. At Finokalia in the eastern Mediterranean, Bougiatioti et al. (2016) derived a hygroscopicity parameter from

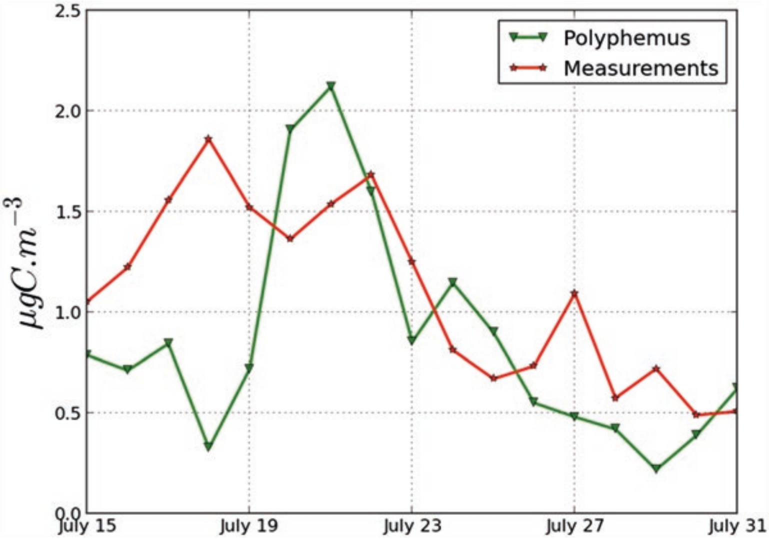

Fig. 5 Submicron WSOC measured (in red) and simulated (in green) at Ersa during the summer of 2013. (Reprinted from Chrit et al. (2017))

CCN and hygroscopic tandem differential mobility analyzer (HTDMA) measurements and linked it to distinct chemical constituents identified with PMF of the chemical constituents measured with an ACSM. Hygroscopicity was found to increase with size, as particles are more atmospherically processed. Furthermore, freshly emitted biomass burning OA was found to be around two to three times less hygroscopic than atmospherically processed biomass burning OA and highly oxidized organic aerosols.

2.2 *Origins and Formation Pathways*

As discussed in the introduction, a large fraction of PM_1 and $PM_{2.5}$ is organic. The study of the properties of OA in the previous sections shed lights on the origins and formation pathways of OA. Note that the interactions between chemical compounds of different origins are strong. For example, in summer over the Mediterranean, Chrit et al. (2018a) simulated a strong sensitivity of OA concentrations to meteorology. This sensitivity is not only because the temperature influences the partitioning between the gaseous and particulate phases but also because the meteorology influences the emissions of biogenic precursors and inorganic sea salt emissions, which are both involved in the formation of OA as discussed below.

Biogenic

In summer, at least 70% of OA was observed to be biogenic at both remote and urban sites. However, the influence of anthropogenic emissions on the formation of biogenic organic aerosols is clearly important, as shown by the modeling work (Sartelet et al., 2012). Freney et al. (2018) showed that less oxidized organic aerosols correlate well with biogenic precursors over forests in the south of France. The contribution of specific signatures for isoprene epoxydiols SOA (IEPOXSOA) was very weak, suggesting that isoprene SOA was not formed through IEPOX.

The modeling studies of Chrit et al. (2017) and Freney et al. (2018) suggest that, at ERSA site and over forests, non-IEPOX SOA from isoprene forms 15–35% of the OA mass, with a large part of OA being formed from the oxidation of monoterpenes (30–50%) and sesquiterpenes (about 10%). A large fraction of oxidation products of biogenic precursors may be hydrophilic, explaining the high fraction of water-soluble carbon observed. The contribution of ELVOCs from monoterpenes was found to be as high as 10%, partly explaining the high oxidation state observed. These compounds may also be involved in new particle formation (NPF), as suggested in the preceding chapter on nucleation (Sellegri & Rose, 2022).

The temperature sensitivity of biogenic SOA (BSOA) formation has been addressed in a modeling study, comparing BSOA under a future climate scenario with different organic aerosol schemes (Cholakian et al., 2019a). Five years of historical simulations and 5 years of future simulations according to a RCP8.5 scenario have been conducted with a modeling chain described in Colette et al. (2013), involving for boundary conditions the IPSL-CM5A-MR global climate model and the LMDz-INCA global CTM, the EURO-CORDEX WRF regional climate simulations, and CHIMERE regional CTM simulations with different organic aerosol schemes. Over Europe and the Mediterranean, BSOA increases for all aerosol schemes as a consequence of increased BVOC emissions triggered by higher temperature. However, a scheme based on the volatility basis set formalism and including functionalization and fragmentation of semi-volatile organic species as well as formation of non-volatile BSOA (Shrivastava et al., 2015) leads to two times higher relative BSOA increases than the molecular standard scheme in CHIMERE without chemical aging (Menut et al., 2013). For the latter scheme, the BVOC increase is partly compensated by increased evaporation of semi-volatile BSOA in a hotter climate, while for the former scheme, this effect is more limited due to the formation of non-volatile BSOA (Cholakian et al., 2019a). Thus, exact knowledge of chemical formation pathways of BSOA is necessary to correctly predict its sensitivity to a future warmer climate.

Marine Aerosol

Satellites (MODIS) show important phytoplankton activity near coasts in the Mediterranean (Chrit et al., 2017) with sea-derived organic particles emitted in the Aitken mode (Schwier et al., 2015, 2017). However, Chrit et al. (2017) showed that the contribution of the sea-origin organic particles to organic concentrations is low (at most 2% in summer) if parameterizations using chlorophyll a (Chl a) as a proxy are used. This agrees with the measurements of Schwier et al. (2017), who found that the correlation between the particle organic fraction and the seawater Chl a was poor in their measurements.

Measurements at Lampedusa found correlations between sulfate and organics (Mallet et al., 2019). For Mallorca and Ersa, the ME2 (Multilinear Engine 2, SoFi toolkit; Canonaco et al., 2013) was used to perform PMF calculation with the whole AMS mass spectra (i.e., including sulfate, nitrate, and ammonium fragments in addition to organic fragments) measured in July 2013 in Mallorca and Ersa. A factor mixing sulfate and oxidized organic fragments (named sulfate-OOA) represents between 15% in Mallorca and 25% in Ersa of the total PM_1 mass. As shown for Mallorca (Fig. 6), the sulfate-OOA factor is, at both sites, extremely well correlated with $CH_3SO_2^+$ fragment, a well-known fragment of methane sulfonic acid (MSA) which is an oxidation of product of dimtehyl sulfide (DMS). Therefore, a significant biogenic marine contribution to the secondary organic aerosol budget is expected.

This biogenic marine contribution may be direct and linked to DMS, as discussed by Sellegri and Rose (Sellegri & Rose, 2022), but it may also be an indirect contribution, for example, by providing an absorbing aqueous mass for condensing non-marine biogenic oxidation products. As shown in Pun et al. (2006) and Couvidat and Seigneur (2011), biogenic oxidation products tend to be strongly hydrophilic. They may condense onto the aqueous phase formed by marine primary inorganic

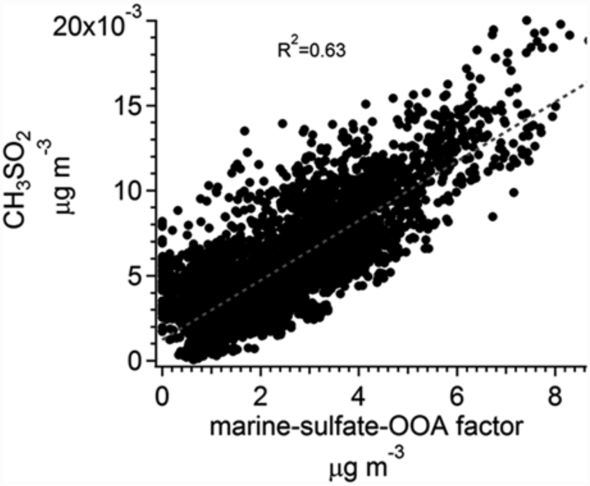

Fig. 6 Correlation, fitted by the dotted line, between the sulfate-OOA factor and the main MSA fragment ($CH_3SO_2^+$) at Mallorca in July 2013. (Courtesy of Nicolas Marchand)

aerosols. In Chrit et al. (2018a), biogenic SOA is formed from the condensation on marine aerosols of hydrophilic oxidation products of monoterpenes, such as pinonaldehyde and pinic acid, and a strong sensitivity of OM_1 concentrations to sea salt emissions is simulated. Other processes may also be involved: the experiments of Wang et al. (2019) showed that particulate sulfate may be formed through the oxidation of dissolved sulfur dioxide by organic peroxides from monoterpene oxidation.

Anthropogenic

The estimate of the anthropogenic contribution to organic aerosols is highly dependent on the quality of the emission inventory. In some parts of the eastern Mediterranean, the anthropogenic contribution is difficult to establish because of difficulties to have accurate emission inventories, as pointed out by Abdallah et al. (2016 and 2018) and Salameh et al. (Salameh et al., 2016; Salameh et al., 2017) for Lebanon and Beirut (see also Borbon et al., 2022). In summer, although the anthropogenic contribution from organic precursors was found to be low, anthropogenic emissions influence the formation of biogenic aerosols by enhancing oxidant concentrations and increasing the absorbing mass (Sartelet et al., 2012). The contribution of anthropogenic precursors, as shown by the ^{14}C measurements, is low in summer. Using modeling, the contribution of single-ring aromatics (toluene, xylene) is simulated to be low (<5%) in both summer and winter (Couvidat et al., 2012; Chrit et al., 2017, 2018b). In summer, I/S-VOCs are found to contribute about 20% or less to OA, while their contribution is major in winter (about 80%). However, other volatile organic compounds emitted from residential wood combustion may strongly influence the OA formation, potentially explaining the high organic oxidation state observed (Stefenelli et al., 2019). At Athens, in the PMF receptor modeling on measured data, Theodosi et al. (2018) estimated that biomass burning is the main source of $PM_{2.5}$ (31%), followed by vehicular emissions (19%) and heavy oil combustion linked to maritime traffic (7%).

Maritime Traffic

Using markers of heavy fuel oil combustion, Becagli et al. (2017) estimated that during summer 2013, the minimum ship emission contributions to PM_{10} were 11% at Lampedusa and 8.6% at Capo Granitola, respectively. Large differences exist in emission inventory of ships (Chrit et al., 2017; Cholakian et al., 2018). However, ships strongly influence OA concentrations over the Mediterranean (Fig. 3). They emit not only primary organic particles and organic precursors but also NO_x leading to an increase of oxidants and hence SOA formation. Organic nitrate is also estimated to be important over the sea during low-wind periods (Chrit et al., 2017).

Wildfires

Although wildfires are sporadic, they can lead to high aerosol loading in the atmosphere. For example, intense wildfire episodes have been reported in Greece and the Balkans (Majdi et al., 2019a). Because wildfires involve large amounts of aerosols, wildfires from other continents, especially the North American continent or Siberia (Brocchi et al., 2018; Guth et al., 2018), impact the Mediterranean domain. Wildfires contribute to high organic aerosol loading (Bougiatioti et al., 2016), with a large contribution of I/S-VOCs (Konovalov et al., 2015; Konovalov et al., 2019; Majdi et al., 2019a). Although their contribution to OA could be as large as 70% during severe fire episodes, volatile organic compounds (phenol, catechol, cresol, xylene, toluene, benzene, naphthalene) may significantly contribute to OA (Majdi et al., 2019b). During wildfires, atmospheric processes of particles affect the hygroscopicity as well as the mixing state of the particles (Bougiatioti et al., 2016).

3 Inorganic Aerosols

Over the Mediterranean, sulfate makes a large fraction of fine aerosols (see introduction and Fig. 2). Sea salt aerosols contain sulfate (Schwier et al., 2015). However, the contribution of sea salt sulfate to PM_1 is small, and sulfate is highly sensitive to the emission inventory (maritime traffic) (Chrit et al., 2018a). Ammonium, sodium, nitrate, and chloride to a lesser extent (Chrit et al., 2018a) are measured to contribute significantly to PM_{10} and PM_1 concentrations. Note that the contribution of sodium is included in the composition depicted in Fig. 2, because ACSM and AMS measurements can only estimate the concentrations of non-refractory compounds.

Sodium and chloride originate mostly from sea salt emission. Although sodium is non-volatile, sea salt chloride evaporates from sea salt aerosols to be replaced by nitrate (Claeys et al., 2017), influencing the gas/particle partitioning of inorganic semi-volatile compounds. The precursors of aerosol nitrate and ammonium are mostly anthropogenic: nitrogen dioxide and ammonia, with a large fraction emitted by traffic and agriculture, respectively. The mixing state of particles influences the gas/particle partitioning. For example, semi-volatile inorganic compounds are strongly influenced by the mixing state of sea salt aerosols. Those may not be well mixed to urban aerosols, with a dynamic exchange between the gas and particle phases (Chrit et al., 2018a; Freney et al., 2018).

Using sensitivity simulations, several studies pointed to the strong influence of maritime traffic, meteorology, and sea salt emissions on inorganic aerosol concentrations in the western Mediterranean region (Cholakian et al., 2018; Chrit et al., 2018a; Guth et al., 2018). Maritime traffic affects inorganic aerosol concentrations, because it emits their precursors (nitrogen dioxide and sulfur dioxide). Meteorology influences the gas-/particle-phase partitioning of semi-volatile inorganics, nitrate, ammonium, and chloride, because of the influence of the temperature. However, temperature influences not only the gas-/particle-phase partitioning but also photochemistry and the formation of semi-volatile compounds, such as inorganic nitrates or organic species (see above) from their precursors, explaining the strong sensitivity of these compounds to climate change (Cholakian et al., 2019b).

4 Conclusions and Recommendations

Organic and inorganic aerosols make a large fraction of fine particles over the Mediterranean. In summer, organics are mostly of biogenic origins, water-soluble, and highly oxidized. Biogenic precursors are monoterpenes, isoprene, and sesquiterpenes. Anthropogenic emissions influence the formation of biogenic organics by influencing the formation of oxidants and by providing an absorbing mass for condensation. The formation processes of biogenic aerosols are fairly well modeled, and 3D air quality models manage to represent the concentrations and properties of OA at remote sites and over forests, despite large uncertainties in biogenic emissions (Jiang et al., 2019). ELVOCs are found to contribute to up to 10% of organics in the western Mediterranean. They may also contribute to the formation and growth of NPF, as suggested in Chapter 6.3. In winter, precursors are mostly anthropogenic, with formation processes that still need to be studied further. The models suggest a strong influence of IS/VOC emissions, whose gas phase is missing in emission inventories. However, as the organic oxidation state is not well modeled in winter, formation processes may need to be revisited, by considering the formation of extremely low volatility organic compounds and organic nitrate from anthropogenic precursors. Note that the viscosity of OA may also influence gas/particle partitioning of organics (Kim et al., 2019).

Among inorganics, sulfate concentrations are especially high, because of ship emissions, as discussed in chapter “Anthropogenic Emissions of Ultrafine Particles in the Mediterranean”. The influence of sulfate sea salt is simulated to be low compared to the influence of ship emissions. Because of the diversity of sources (anthropogenic, sea salt, biogenic) and the high photochemistry in summer, an accurate representation of gas/particle partitioning is required to represent inorganic concentrations in summer, with a large influence of thermodynamic and aerosol mixing state. Primary sea salt organic emissions are also estimated to have a low contribution to organics. However, sea salt emissions strongly enhance the formation of organic aerosols by providing an absorbing aqueous mass to the hydrophilic semi-volatile organics formed from the oxidation of biogenic precursors. Inorganics provide not only an aqueous absorbing mass, but their interactions with organics need to be better understood and modeled, e.g., deviation from ideal thermodynamic behavior as represented by activity coefficients is important.

The concentrations of secondary organic aerosols have been simulated to increase in the Mediterranean in 2050 because of climate change. Increased temperatures lead to an increase in biogenic VOC emissions and to a shift of gas-particulate phase equilibrium toward gas phase. The increase in biogenic emissions was shown to be the most impacting factor, explaining the increase of secondary organic aerosols. Over cities, the increase of secondary aerosols may also be due to an increase in oxidant concentrations, because of NO_x emission reductions. Land-use changes could also affect these NO_x and biogenic emissions and should be taken into account in future simulations.

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