



1 **Implementation of the ORACLE (v1.0) organic** 2 **aerosol composition and evolution module into the** 3 **EC-Earth3-AerChem model**

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18
19 **Abstract.** Simulating the composition and evolution of organic aerosol (OA) in Earth
20 System Models (ESMs) presents significant challenges due to the high computational
21 demands of detailed chemical mechanisms. The computationally efficient ORACLE
22 module employs the volatility basis set framework and can simulate secondary
23 organic aerosol (SOA) formation from a range of precursors, including volatile
24 (VOCs), intermediate-volatility (IVOCs), semi-volatile (SVOCs), and low-volatility
25 organic compounds (LVOCs). In this study, a lite configuration of the ORACLE v1.0
26 module (ORACLE-lite) is implemented into the TM5-MP global chemical transport
27 model (CTM), which represents the chemistry-transport component of the EC-Earth3-
28 AerChem ESM. SOA formation from anthropogenic VOCs is neglected to reduce the
29 number of surrogate species and further improve computational efficiency. For the
30 standalone TM5-MP simulation, the global annual mean surface total OA
31 concentration using ORACLE-lite is approximately $1.1 \mu\text{g m}^{-3}$, representing a 25%
32 increase compared to the previous version of the model. The annual atmospheric OA
33 burden also increases by 50%, reaching 3.67 Tg. Corresponding predictions from EC-
34 Earth3-AerChem are slightly higher, with a surface total OA concentration of 1.16
35 $\mu\text{g m}^{-3}$ and an atmospheric burden of 3.83 Tg, representing increases of 30% and



36 60%, respectively, compared to the previous version of the model. Comparison of
37 monthly measured PM_{2.5} OA concentrations from Europe and the US with the
38 corresponding predictions shows that the models bias is reduced by approximately
39 half in the standalone TM5-MP simulation and by a factor of three in EC-Earth3-
40 AerChem when ORACLE-lite is implemented. These enhancements enable more
41 accurate and computationally feasible assessments of the climate impacts of
42 individual organic aerosol components in future ESM studies.

43

44 **1. Introduction**

45 Atmospheric particulate matter (PM) not only affects air quality and human health but
46 also has significant implications for the climate (Monks et al., 2009; Shrivastava et
47 al., 2017; Zhang et al., 2020). Organic aerosol (OA) is a major component of PM
48 contributing between 20% and 90% to the total aerosol mass (Kanakidou et al., 2005;
49 Zhang et al., 2007; Tsimpidi et al., 2025). Since anthropogenic carbonaceous
50 emissions are significant contributors to climate forcing and air pollution (IPCC,
51 2021), it is important to simulate the chemical composition and evolution of OA in
52 Earth System Models (ESMs). This will reduce uncertainties related to aerosols and
53 improve climate predictions.

54 Organic mass is categorized into primary (POA) and secondary organic
55 aerosol (SOA) based on its formation mechanism. Particulate organic mass that is
56 directly emitted into the atmosphere from various sources is referred to as POA. In
57 contrast, SOA is produced in the atmosphere through the oxidation of gas-phase
58 organic compounds. SOA is often the dominant component of OA (Zhang et al.,
59 2007; Crippa et al., 2013; Hu et al., 2016; Nault et al., 2018). However, SOA
60 concentrations are often underestimated in global climate and chemical transport
61 models (Heald et al., 2005; Tsigaridis et al., 2014; Tsimpidi et al., 2016; Bergman et
62 al., 2022). This is partially due to neglected processes in the models (Robinson et al.,
63 2007), such as the evaporation of POA, the oxidation of the resulting vapors in the gas
64 phase and their subsequent condensation into the particle phase, SOA formation from
65 intermediate-volatility organic compounds (IVOCs) and the chemical aging of
66 volatile organic compounds (VOCs). Many studies have shown that the
67 photooxidation of emissions from fossil fuel combustion and biomass burning can
68 lead to the formation of significant SOA concentrations (Kroll and Seinfeld, 2008;



69 Grieshop et al., 2009; Hennigan et al., 2011; Tsimpidi et al., 2017; Ma et al., 2018;
 70 Lim et al., 2019; Fang et al., 2021), which is often not accounted for in many ESMs.

71 Climate models usually treat POA and SOA as non-volatile and non-reactive
 72 particles that are directly emitted into the atmosphere (Kanakidou et al., 2005;
 73 Tsimpidi et al., 2014; Pai et al., 2020). Donahue et al. (2006) introduced the volatility
 74 basis set (VBS) framework to capture the changes in OA volatility. This framework
 75 describes the partitioning of OA, assuming it is semi-volatile and photochemically
 76 reactive and that it is distributed across logarithmically spaced volatility bins. By
 77 using this approach, both the emissions of intermediate and semi-volatile primary
 78 aerosols, as well as SOA formation and its aging processes can be simulated. This
 79 approach has already been implemented in several regional and global chemical
 80 transport models (Tsimpidi et al., 2010; Jathar et al., 2011; Shrivastava et al., 2011;
 81 Bergström et al., 2012; Woody et al., 2016; Chen et al., 2019; Jiang et al., 2019) and
 82 in a few ESMs (Gao et al., 2024; Irfan et al., 2024). Many of these modeling studies
 83 have demonstrated improved predictions of OA concentrations by incorporating the
 84 VBS framework into their simulations. However, its implementation in large-scale
 85 models, such as ESMs, remains limited due to its high computational expense.

86 Tsimpidi et al. (2014) developed the ORACLE module, which is based on the
 87 VBS framework, and implemented it in the ECHAM/MESSy Atmospheric Chemistry
 88 (EMAC) model (Jöckel et al., 2006). ORACLE reduces the computational cost by
 89 utilizing a small number of surrogate OA species by employing a novel lumping
 90 method. However, the 92 species used in the full configuration of the ORACLE
 91 module to describe OA and its volatility are still excessive for ESM simulations
 92 increasing significantly the computational cost. To address the computational
 93 constraints of ESMs, Tsimpidi et al. (2025) introduced a lite configuration of the
 94 ORACLE module (hereafter ORACLE-lite), further reducing the number of species
 95 used to describe OA and its volatility from 92 to 18. Although simplifications were
 96 made to reduce the computational cost, ORACLE-lite continues to effectively
 97 simulate the contributions of low volatility organic compounds (LVOCs), semi-
 98 volatile organic compounds (SVOCs), IVOCs, and VOCs to SOA formation
 99 (Tsimpidi et al., 2025). ORACLE-lite is recommended for ESM simulations (Riipinen
 100 et al., 2025).

101 The aim of this study is to incorporate a computationally efficient OA
 102 volatility scheme based on ORACLE-lite into the chemistry-transport component of



103 EC-Earth3-AerChem ESM to simulate OA concentrations, composition, and
 104 evolution. Section 2 provides an overview of the model, focusing mostly on the new
 105 implementations. In particular, we describe the version of the EC-Earth ESM used,
 106 the implemented OA volatility scheme, and the conducted simulations. In Section 3,
 107 we present the model-derived OA atmospheric concentrations and their evaluation
 108 with available observations. Finally, in Section. 4, we discuss the impact of the VBS
 109 framework on the simulated OA atmospheric concentrations and we summarize the
 110 global implications of explicitly representing POA emissions in a climate–chemistry
 111 model, along with the plans for future model development.

112

113 **2. Model description**

114 **2.1 The EC-Earth3 Earth System Model**

115 The EC-Earth3-AerChem configuration (EC-Earth3-AerChem version 3.5.0) of the
 116 EC-Earth3 (Döscher et al., 2021) has been used for this work. EC-Earth3 contributed
 117 to Phase 6 of the Coupled Model Intercomparison Project (CMIP6; Eyring et al.,
 118 2016). Its atmospheric general circulation model (GCM) is based on cycle 36r4 of the
 119 Integrated Forecast System (IFS), from the European Centre for Medium-Range
 120 Weather Forecasts (ECMWF), which includes the H-TESEL land surface model
 121 (Balsamo et al., 2009). The ocean model is the version 3.6 of the Nucleus for
 122 European Modelling of the Ocean (NEMO; Rousset et al., 2015), with sea ice
 123 processes represented by the Louvain-la-Neuve sea ice model (LIM; Vancoppenolle
 124 et al., 2009; Rousset et al., 2015). The majority of information exchange and
 125 interpolation between modules is managed by the Ocean Atmosphere Sea Ice Soil
 126 coupler, version 3 (OASIS3; Craig et al., 2017). EC-Earth3-AerChem includes TM5-
 127 MP (Tracer Model 5, Massively Parallel version; Kroll et al., 2005; Huijnen et al.,
 128 2010; van Noije et al., 2014; Williams et al., 2017) for the simulation of aerosols and
 129 atmospheric chemistry. TM5-MP can be also used as a standalone CTM driven by
 130 offline meteorological and surface fields from the ERA-Interim reanalysis, developed
 131 by the European Centre for Medium-Range Weather Forecasts (ECMWF; Dee et al.,
 132 2011). It simulates the atmospheric life cycle of air pollutants, including emissions,
 133 advection, convection, vertical diffusion, and removal by dry and wet deposition, as
 134 well as chemical and microphysical transformations. Gas-phase chemistry is
 135 described by mCB05, a modified version of the CB05 carbon bond mechanism
 136 (Yarwood et al., 2005; Williams et al., 2017). For the gas and particle equilibrium



calculations of $\text{NH}_3/\text{NH}_4^+$ and $\text{HNO}_3/\text{NO}_3^-$, the ISORROPIA-lite model is used (Kakavas et al., 2022) neglecting the effect of organic aerosol water on inorganic aerosol thermodynamics. The organic aerosol water contribution to the total aerosol water is calculated separately, based on Myriokefalitakis et al. (2022). To simulate the composition and evolution of OA the ORACLE-lite module (Tsimpidi et al., 2025) is used.

The aerosol population and its evolution are treated by the modal two-moment aerosol model M7 (Vignati et al., 2004). M7 includes four water soluble modes (nucleation, Aitken, accumulation, and coarse) and three insoluble modes (Aitken, accumulation, and coarse). The dry radius size ranges for the aerosol modes are defined as follows: nucleation mode ($r_p < 5$ nm), Aitken mode ($5 < r_p < 50$ nm), accumulation mode ($50 < r_p < 500$ nm), and coarse mode ($r_p > 500$ nm). Particles within each mode are assumed to be internally mixed. Each mode follows a lognormal size distribution with a fixed geometric standard deviation. The M7 model tracks the evolution of both total particle number and the mass of each species within each mode. In this work, we incorporated both POA and SOA into the default soluble POA modes (Aitken, accumulation, and coarse) of the M7 model to track aerosol number distribution. The existing M7 species also include SOA formed from biogenic VOCs (Bergman et al., 2022), along with sulfate, black carbon, sea salt and dust. Additionally, TM5-MP simulates the concentrations of nitrate, ammonium, and methane sulfonic acid using a bulk aerosol approach. More details about TM5-MP and EC-Earth3-AerChem can be found in van Noije et al. (2021) and Myriokefalitakis et al. (2022).

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161 2.2 The ORACLE module

ORACLE (v1.0) uses fixed, logarithmically spaced saturation concentration bins and assumes bulk equilibrium between the gas and particle phases. The OA mass is then distributed among the size modes (Aitken soluble, accumulation soluble, and coarse soluble) following Pandis et al. (1993), using the dry radius of each size mode from the M7 model. ORACLE simulates: (i) the partitioning of POA from LVOC emissions, (ii) the partitioning of POA from SVOC emissions and gas-phase oxidation of the remaining vapors, followed by their condensation into the particle phase to form SOA, and (iii) the gas-phase oxidation of IVOC and VOC emissions and the subsequent condensation of the oxidation products to form SOA. The volatility bins



are defined by saturation concentration (C^*) ranges of 10^{-2} to $10^{-1} \mu\text{g m}^{-3}$ for LVOCs, 10^0 to $10^2 \mu\text{g m}^{-3}$ for SVOCs, 10^3 to $10^6 \mu\text{g m}^{-3}$ for IVOCs, and $>10^6 \mu\text{g m}^{-3}$ for VOCs.

To further reduce computational expense, we implemented a lite configuration of the ORACLE module (hereafter ORACLE-lite) in the TM5-MP model, which represents the chemistry-transport component of the EC-Earth3-AerChem ESM by introducing a reduced set of surrogate species. This includes POA and primary organic gas (POG) for LVOC emissions, POA and POG for SVOC emissions, and POG for IVOC emissions, as well as SOA and secondary organic gas (SOG) formed through the oxidation of SVOC and IVOC emissions by hydroxyl radicals. Note however, that the SOA formation from biogenic VOC emissions (isoprene and monoterpenes) is already represented in the models, as described by Bergman et al. (2022), while SOA formation from anthropogenic VOC emissions is neglected. As a result, the number of surrogate species used to represent OA and its volatility in ORACLE-lite was reduced from 18 to 9. An overview of the characteristics of the lite configuration of the ORACLE module used in this study is shown in Table 1.

2.3 Simulations

In this study, present-day simulations were performed using atmosphere-only runs of EC-Earth3-AerChem (hereafter referred to as EC-Earth) for the years 2000-2010. In addition, standalone simulations with TM5-MP for the year 2005 driven by ERA-Interim (Dee et al., 2011) were performed. In the EC-Earth simulation, TM5-MP is coupled to the IFS atmospheric dynamics. Sea surface temperature and sea ice concentration fields were prescribed using input files provided through the AMIP interface (Taylor et al., 2000). Consequently, the atmospheric and chemistry modules follow the standard EC-Earth3-AerChem configuration used in CMIP6. The IFS component is configured with a horizontal resolution of T255 (approximately 80 km), 91 vertical levels extending up to 0.01 hPa, and a time step of 45 minutes. TM5-MP, in both its standalone and EC-Earth configurations, uses a horizontal resolution of $3^\circ \times 2^\circ$ (longitude $\times$ latitude) and 34 vertical levels extending up to 0.1 hPa (~ 60 km).

For this work, two types of simulations were performed for both TM5-MP and EC-Earth: (a) using the default OA representation and emissions, in which OA is treated as non-volatile, non-reactive and emitted exclusively as POA, and (b)



204 incorporating the ORACLE-lite module with modified emissions (hereafter referred
 205 to as VBS).

206 In the VBS configuration, the emission factors used to distribute traditional
 207 POA emissions into LVOCs and SVOCs are based on the work of Tsimpidi et al.
 208 (2025). For fossil fuel combustion sources, emission factors of 0.09 and 0.71 are
 209 assigned to LVOCs and SVOCs, respectively. For biomass burning, the
 210 corresponding factors are 0.2 for LVOCs and 0.5 for SVOCs. These emissions are
 211 assigned to volatility bins with C^* of $10^{-2} \mu\text{g m}^{-3}$ and $10^1 \mu\text{g m}^{-3}$ for LVOCs and
 212 SVOCs, respectively (Table 1). Please note that IVOCs exist almost exclusively in the
 213 gas-phase (Pandis et al., 2013) and are not fully accounted for in traditional emission
 214 inventories, despite their potentially substantial role in SOA formation. Previous
 215 studies estimate IVOCs emissions to range from 0.25 to 2.8 times those of traditional
 216 POA emissions (Schauer et al., 2002). In this study, we assume that IVOCs emissions
 217 are equal to 0.3 times the traditional POA emissions for biomass burning and 1.7
 218 times for fossil fuel combustion sources, following the estimates of Tsimpidi et al.
 219 (2025). These emissions are assigned to the volatility bin with $C^* = 10^4 \mu\text{g m}^{-3}$.
 220 Overall, LVOCs and SVOCs are assumed to be initially emitted in the particle phase
 221 as POA, while IVOCs are emitted solely in the gas-phase.

222

223 2.4 Observations

224 To evaluate the impact of the VBS scheme on simulated aerosol concentrations, we
 225 compared the models results with monthly surface-level observations of $\text{PM}_{2.5}$ OA
 226 concentrations during 2005. We used data from two freely available observational
 227 networks: the United States Interagency Monitoring of Protected Visual
 228 Environments (IMPROVE; <https://views.cira.colostate.edu/fed/QueryWizard>, last
 229 access: 2 June 2025) and the European Monitoring and Evaluation Project (EMEP;
 230 <https://ebas-data.nilu.no/Default.aspx>, last access: 2 June 2025). For the IMPROVE
 231 network, we used monthly OA concentrations from 174 stations while for EMEP, data
 232 were available from only 3 stations for the simulated period. Please note that both
 233 IMPROVE and EMEP networks measure particulate organic carbon (OC) instead of
 234 total organic mass in the particles. To convert OC to organic mass, we applied a
 235 constant factor. For the IMPROVE network, the suggested factor equals to 1.8
 236 (Pitchford et al., 2007). For EMEP, we followed the IMPROVE network



237 recommendation and assumed a factor of 1.8 for EMEP stations to maintain
 238 consistency.

239

240 **2.5 Models performance**

241 To evaluate the models performance, specific statistical metrics were calculated for
 242 both configurations of TM5-MP and EC-Earth simulations. These include mean bias
 243 (MB), mean absolute gross error (MAGE), normalized mean bias (NMB), normalized
 244 mean error (NME), and root-mean-square error (RMSE) defined as follows:

$$MB = \frac{1}{N} \sum_{i=1}^N (P_i - O_i) \quad (1)$$

$$MAGE = \frac{1}{N} \sum_{i=1}^N |P_i - O_i| \quad (2)$$

$$NMB = \frac{\sum_{i=1}^N (P_i - O_i)}{\sum_{i=1}^N O_i} \quad (3)$$

$$NME = \frac{\sum_{i=1}^N |P_i - O_i|}{\sum_{i=1}^N O_i} \quad (4)$$

$$RMSE = \left[\frac{1}{N} \sum_{i=1}^N (P_i - O_i)^2 \right]^{1/2} \quad (5)$$

245 where P_i is the predicted OA concentration, O_i is the observed OA value at the same
 246 monthly averaged time, and N is the total number of measurements used for the
 247 comparison.

248 NME (in %) and MAGE (in $\mu\text{g m}^{-3}$) measure the total difference between
 249 model predictions and observations, including both bias and scatter. In contrast, NMB
 250 (in %) and MB (in $\mu\text{g m}^{-3}$) specifically reflect systematic errors. RMSE (in $\mu\text{g m}^{-3}$)
 251 combines both the variability (scatter) and bias of the predictions into a single metric.
 252 Because NME and MAGE include bias effects, their values are typically equal to or
 253 greater than those of NMB and MB. When NME and NMB (or MAGE and MB) are
 254 similar in magnitude, most of the error is due to bias. However, if NME and MAGE
 255 are substantially larger than NMB and MB, this indicates that scatter also contributes
 256 significantly to the discrepancy between predictions and observations.

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258

259



2.6 Emissions

The annual present-day emissions used in the chemistry transport component of EC-Earth (TM5-MP) with the VBS configuration are shown in Fig. 1. In the default OA configuration, particulate organic matter is emitted exclusively as POA (Fig. S1; 52.4 Tg yr⁻¹) and is assumed to have constant carbon content, expressed as the ratio of total OA mass to the mass of the carbon it contains. This ratio is used to convert POA emissions, typically expressed as organic carbon (OC) mass to OA mass. In this study, a ratio of 1.6 is applied across all POA sources based on previous works (Turpin and Lim, 2001; Reid et al., 2005; Aiken et al., 2008; van Noije et al., 2021). In the VBS configuration, emissions are distributed into three volatility bins based on the emission factors described in Section 2: LVOCs (7.6 Tg yr⁻¹), SVOCs (31.8 Tg yr⁻¹), and IVOCs (53.1 Tg yr⁻¹) (see also Table 2). LVOCs and SVOCs represent POA emissions, which are lower in total than in the default OA configuration because a portion of the traditional organic mass emissions is reassigned to IVOCs. The corresponding annual mean emissions used for the EC-Earth simulation are shown in Fig. 2. The emissions are higher in regions such as China, India, Bangladesh, southern Africa, and South America. Emissions from shipping are also present over oceanic regions.

3. Results

3.1 Budget calculations

This section presents the global budgets, atmospheric burdens, and lifetimes of OA components from EC-Earth and standalone TM5-MP simulations during 2005 using the VBS configuration (Table 1). For completeness, SOA from biogenic VOCs (bSOA-v) is also included.

In the standalone TM5-MP simulation for 2005, SOA production from SVOCs (SOA-sv) and IVOCs (SOA-iv) is 19.83 Tg yr⁻¹ and 37.02 Tg yr⁻¹, respectively. The total annual SOA production is 109.19 Tg yr⁻¹. This value falls within the range of 50-380 Tg yr⁻¹ of Spracklen et al. (2011) and is close to their best estimate of 140 Tg yr⁻¹. The relative contributions to the annual SOA production are 18.2% from SVOCs, 33.9% from IVOCs, and 47.9% from biogenic VOCs. Anthropogenic SOA production is higher near source regions (Fig. 3), with hotspots in South America, southern Africa, India, Bangladesh, and China. Seasonally, the production of SOA-sv is higher in summer (Fig. S2), whereas SOA-iv production peaks in winter, especially



294 over India, China, and Central Africa (Fig. S3). This seasonal trend will be discussed
 295 further in the next section.

296 In the EC-Earth simulation, the annual mean production for 2000-2010 of
 297 SOA-sv and SOA-iv is $19.62 \pm 1.67 \text{ Tg yr}^{-1}$ and $38.28 \pm 7.32 \text{ Tg yr}^{-1}$, respectively
 298 (Table 2; Fig. 3), with a total SOA production of $109.22 \pm 10.23 \text{ Tg yr}^{-1}$. The
 299 contributions to total SOA production are 18% from SVOCs, 35% from IVOCs, and
 300 47% from biogenic VOCs. Annual (Fig. S4) and seasonal (Figs. S5 and S6) SOA
 301 production indicate no significant differences between the TM5-MP and EC-Earth
 302 simulations during 2005. Minor discrepancies arise from differences in meteorology,
 303 which is predicted in EC-Earth but prescribed from reanalysis in the TM5-MP
 304 simulation.

305 The contributions to the annual atmospheric burden of total OA (3.67 Tg) in
 306 the standalone TM5-MP simulation are 18.7% from POA, 16.4% from SOA-sv,
 307 32.9% from SOA-iv, and 32% from bSOA-v. Compared to the default simulation, in
 308 which SOA is produced only from biogenic VOCs and all anthropogenic OA is
 309 treated as POA, the annual atmospheric burden of total OA increased by
 310 approximately 50% in the VBS simulation. In EC-Earth, the annual mean atmospheric
 311 burden using the VBS configuration is 60% higher than in the default configuration,
 312 reaching $3.83 \pm 0.21 \text{ Tg}$. The respective contributions are 19.3% from POA, 15.9%
 313 from SOA-sv, 34% from SOA-iv, and 30.8% from bSOA-v.

314

315 **3.2 Atmospheric concentrations**

316 The annual mean surface concentrations of POA, SOA-sv, SOA-iv, and bSOA-v in
 317 the standalone TM5-MP simulation with the VBS configuration are shown in Fig. 4.
 318 POA levels are higher than those of SOA-sv and SOA-iv especially in regions like
 319 India and China with higher LVOC and SVOC emissions. Our simulations indicate
 320 that the emitted POA undergoes evaporation and is subsequently oxidized by
 321 hydroxyl radicals in the gas phase, leading to the formation of SOA-sv through re-
 322 condensation. This is consistent with recent experimental studies especially for
 323 biomass burning emissions (e.g., Sengupta et al., 2020; Fang et al., 2021). Biomass
 324 burning emissions from residential heating are typically higher during winter, and the
 325 lower temperatures enhance partitioning to the particle phase, leading to increased



326 POA concentrations, especially in regions such as China, Bangladesh, Central Africa
 327 and India (Fig. S7).

328 The annual mean concentrations of SOA-sv are lower than those of POA, as
 329 only a fraction of POA evaporates, undergoes photooxidation, and subsequently
 330 condenses into the particle phase. The oxidation of IVOCs, producing lower-volatility
 331 products, also contributes to SOA-sv formation. Please note that in ORACLE-lite, the
 332 volatility bin representing SOA-sv corresponds to a C^* value of $10^{-2} \mu\text{g m}^{-3}$,
 333 indicating low volatility and a predominant presence in the particle phase. The higher
 334 SOA-sv concentrations predicted during summer compared to winter (Table S1; Fig.
 335 S8) are due to the higher summer temperatures promoting POA evaporation, and the
 336 increased sunlight which enhances subsequent photooxidation and formation of SOA.

337 Higher annual mean concentrations of SOA-iv compared to SOA-sv are
 338 predicted, primarily due to the higher emissions of IVOCs (Table 1; Fig. 1) and the
 339 different formation mechanism. IVOCs can directly undergo oxidation by hydroxyl
 340 radicals, producing lower-volatility products that subsequently condense into the
 341 particle phase. Predicted SOA-iv concentrations are higher in winter than in summer
 342 (Table S1), particularly in regions such as China, India, Bangladesh, and Europe (Fig.
 343 S9). This is attributed to increased biomass burning emissions in these regions and
 344 lower temperatures, which enhance partitioning of the semi-volatile OA components
 345 into the particle phase. However, in regions such as South America and southern
 346 Africa, where major rainforests like the Amazon and Congo Basin are located, SOA-
 347 iv concentrations are higher during summer due to wildfires.

348 The annual mean surface concentrations of total OA in the standalone TM5-
 349 MP simulation with the VBS configuration are shown in Fig. 5. Higher concentrations
 350 are predicted in regions with higher precursor emissions, while as altitude increases
 351 the concentrations of OA decrease as expected, with higher concentrations between
 352 15°S and 45°N (Fig. 5b). At higher altitudes, SOA concentrations are higher
 353 compared to POA, because organic gases can be efficiently transported upward and
 354 oxidized, leading to the formation of lower-volatility SOA (Fig. S10). Additionally,
 355 SOA formed at these altitudes tends to have a longer atmospheric lifetime, as it is less
 356 affected by wet and dry deposition processes (Tsimpidi et al., 2014).

357 The VBS configuration predicts significantly higher annual mean total OA
 358 concentrations (by up to 100%) compared to the default TM5-MP configuration,
 359 particularly in regions such as India, China, and northern Africa (Fig. 5c). Significant



360 increases are also predicted over oceanic regions, including the Indian, Atlantic and
 361 Pacific Oceans. In addition to S/IVOC emissions from shipping (Fig. 1), this increase
 362 is largely driven by the long-range transport of IVOCs, which contributes to SOA-iv
 363 formation far from emission sources (Aiken et al., 2009; Hildebrandt et al., 2010).
 364 This is further supported by the higher increases in total OA concentrations predicted
 365 in these regions during winter compared to summer (Fig. S11), attributed to higher
 366 SOA-iv levels in the colder season (Fig. S9). At higher altitudes, the VBS
 367 configuration in general predicts higher OA concentrations than the default
 368 configuration, particularly between 0° and 45°N (Fig. 5d). However, in the uppermost
 369 levels of the model, the default configuration predicts higher OA concentrations.
 370 Nevertheless, in both simulations, these values are extremely low (below 0.001 $\mu\text{g m}^{-3}$)
 371 m^{-3}), rendering the absolute differences negligible.

372 The annual mean surface concentrations of POA and SOA in the EC-Earth
 373 simulation with the VBS configuration for 2000-2010 are shown in Fig. 6. Similar to
 374 the TM5-MP simulation, higher SOA concentrations than POA are predicted. With
 375 increasing altitude, SOA concentrations remain higher than POA because organic
 376 gases are efficiently transported upward and oxidized, producing lower-volatility
 377 SOA. However, compared to the TM5-MP simulation, there are some differences in
 378 the global distribution of the annual mean surface total OA for 2005 (Fig. S12). More
 379 specifically, in regions such as South America, Africa, India, and China, EC-Earth
 380 predicts higher total OA concentrations (up to 4 $\mu\text{g m}^{-3}$) during 2005 due to the higher
 381 production of SOA-iv. There are also regions such as Europe in which TM5-MP
 382 predicts higher total OA concentrations than EC-Earth. At higher altitudes, TM5-MP
 383 in general predicts higher OA concentrations than EC-Earth, except in the region
 384 between 5° S and 10°N up to 600 hPa (Fig. S12). However, in all cases, the
 385 differences are lower than 0.5 $\mu\text{g m}^{-3}$. These discrepancies stem from differences in
 386 meteorology since EC-Earth uses meteorology predicted by IFS, while TM5-MP
 387 relies on prescribed reanalysis data. More specifically, in these regions, either lower
 388 predicted temperatures or lower precipitation rates in EC-Earth affect OA
 389 concentrations through partitioning and deposition, respectively (Fig. S13).

390 Overall, higher concentrations of OA are predicted by both models in regions
 391 such as India, South America, southern Africa, and China, where precursor emission
 392 levels are higher. The annual global mean surface concentration of total OA in the
 393 TM5-MP simulation using the VBS configuration is 1.07 $\mu\text{g m}^{-3}$, corresponding to an



394 increase of 25% relative to the default configuration. In EC-Earth, the corresponding
 395 annual global mean surface concentration is $1.16 \mu\text{g m}^{-3}$, representing an increase of
 396 30% relative to the default configuration. The contributions of individual OA
 397 components to the annual global mean surface concentration of OA are 29.9% from
 398 POA, 13.1% from SOA-sv, 29% from biogenic SOA, and 28% from SOA-iv. This
 399 highlights the substantial role of IVOCs in contributing to total OA, despite their
 400 omission from traditional emission inventories. Additionally, our simulations indicate
 401 that temperature influences the partitioning of oxidized IVOC products into the
 402 particle phase, with lower temperatures favoring this process. In contrast, oxidized
 403 SVOC products are treated as low-volatility compounds in the ORACLE-lite module
 404 and predominantly remain in the particle phase under typical atmospheric conditions.

405

406 **3.3 Models evaluation**

407 Figure 7 shows the comparison between predicted $\text{PM}_{2.5}$ OA concentrations and
 408 corresponding measurements for both of TM5-MP and EC-Earth simulations during
 409 2005. Each point on the scatterplot represents a monthly average value at a specific
 410 monitoring station. Compared to the default configuration, the VBS configuration of
 411 TM5-MP predicts higher OA concentrations at all stations, with model results
 412 generally falling closer to the 1:1 line. More specifically, in both TM5-MP
 413 simulations, OA concentrations are generally underpredicted at the examined stations,
 414 as indicated by negative MB and NMB values (Table 3). However, the VBS
 415 configuration reduces this underprediction by approximately half (MB = $-0.28 \mu\text{g m}^{-3}$,
 416 NMB = -13.2%) compared to the default configuration (MB = $-0.57 \mu\text{g m}^{-3}$,
 417 NMB = -27.1%). Additionally, both NME and RMSE values, which are relatively
 418 low in the default simulation (NME = 42%, RMSE = $1.57 \mu\text{g m}^{-3}$), improve further in
 419 the VBS simulation (NME = 38.9%, RMSE = $1.5 \mu\text{g m}^{-3}$), indicating reduced scatter.
 420 The same applies for MAGE, which decreased from $0.89 \mu\text{g m}^{-3}$ in the default
 421 simulation to $0.82 \mu\text{g m}^{-3}$ in the VBS simulation. The corresponding EC-Earth
 422 metrics for 2005 for both configurations are also shown in Table 3. EC-Earth also
 423 underpredicts OA concentrations at the examined stations. However, the VBS
 424 configuration of EC-Earth reduces the underprediction by approximately a factor of
 425 three (MB = $-0.17 \mu\text{g m}^{-3}$, NMB = -8.1%) compared to the default configuration
 426 (MB = $-0.54 \mu\text{g m}^{-3}$, NMB = -25.7%). Compared to the standalone TM5-MP



simulation with the VBS configuration, MB and NMB are lower, whereas MAGE and NME are higher ($MAGE = 0.94 \mu\text{g m}^{-3}$, $NME = 44.5\%$). This mainly indicates that the additional SOA-iv production in EC-Earth, resulting from differences in meteorological treatment, further reduces systematic errors but increases bias and scatter.

Figure 8 shows the annual cycle of monthly mean $\text{PM}_{2.5}$ OA concentrations at EMEP and IMPROVE sites in the TM5-MP offline and EC-Earth simulations during 2005. For TM5-MP, the VBS configuration predicts higher concentrations throughout the year compared to the default configuration. However, $\text{PM}_{2.5}$ OA is still underpredicted, particularly at European sites and during winter. This underestimation may result from the omission of oxidation of biomass burning emissions by NO_3 radicals, as well as uncertainties in the biomass burning emissions (Reddington et al., 2019; Hua et al., 2024). The same applies for EC-Earth simulations.

Despite uncertainties in the emissions for fuel combustion and biomass burning, the predictions of OA concentrations using the VBS configuration show improved performance and are generally in good agreement with measurements. Please note that the formation of SOA from anthropogenic VOC emissions (to reduce computational cost) and from oxidation by NO_3 radicals is neglected in the model, which may partially explain the remaining bias. The previous study of Tsimpidi et al. (2014) indicated that SOA from anthropogenic VOCs contributes only about 15% to the global average surface OA concentration. Additionally, the absence of biogenic SOA formation in the models from sesquiterpenes may also contribute to the underprediction (Bergman et al., 2022; Dada et al., 2023).

4. Summary and conclusions

We have implemented a lite configuration of the ORACLE module into the TM5-MP CTM, which represents the chemistry-transport component of the EC-Earth3-AerChem ESM. This version of the module applies the VBS framework to simulate SOA formation from LVOCs, SVOCs, and IVOCs.

The incorporation of ORACLE-lite significantly improved the representation of OA formation and atmospheric behavior both in the standalone TM5-MP and the EC-Earth. The models evaluation against monthly measured $\text{PM}_{2.5}$ OA concentrations from Europe (EMEP) and US (IMPROVE) stations indicated that OA concentrations were generally underpredicted. However, the VBS configuration reduced NMB by



461 nearly half in TM5-MP and a factor of three in EC-Earth and improved the overall
 462 agreement. There is a remaining NMB in both models (−13% in TM5-MP and −8% in
 463 EC-Earth) which can be explained by the absence of SOA formation from
 464 anthropogenic VOC emissions and sesquiterpenes or via oxidation by NO₃ radicals,
 465 which can be subjects for future work. Compared to the traditional POA (default)
 466 configuration, the VBS implementation increased the global annual mean surface OA
 467 concentration in TM5-MP by 25% to 1.07 μg m^{−3} and the atmospheric OA burden by
 468 50%, to 3.67 Tg. Corresponding predictions from EC-Earth were slightly higher, with
 469 a surface OA concentration of 1.16 μg m^{−3} and an atmospheric burden of 3.83 Tg,
 470 representing increases of 30% and 60%, respectively. These changes resulted
 471 primarily from the inclusion of SOA production from S/IVOCs and the treatment of
 472 gas-particle partitioning and chemical aging, processes absent in the default OA
 473 scheme.

474 Our results indicate that SOA is the dominant contributor to total OA surface
 475 concentrations and atmospheric burden, whereas POA contributes less than 30% to
 476 both, highlighting the importance of including gas-phase oxidation and partitioning of
 477 OA in ESMs. The seasonal and spatial variability of SOA was also better captured,
 478 with higher concentrations predicted in regions with intense biomass burning and
 479 anthropogenic activity, such as India, China, and sub-Saharan Africa.

480 Overall, the lite configuration of ORACLE module captures well the key
 481 processes driving OA formation and evolution, offering a more realistic simulation of
 482 OA concentrations without significantly increasing computational cost
 483 (approximately 8%). This efficient and robust configuration supports future studies on
 484 the climatic impacts of OA within ESMs.

485

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498

499 **Code availability**

500 The EC-Earth3-AerChem code (version 3.5.0) is available to members of the EC-
501 Earth consortium via the EC-Earth development portal (<https://dev.ec-earth.org/>, last
502 accessed: 12 November 2025). Model components developed at ECMWF, such as the
503 IFS atmospheric model, are the intellectual property of ECMWF and its member
504 states. Access to the EC-Earth3-AerChem version 3.5.0 source code can be requested
505 through the EC-Earth website (<http://www.ec-earth.org/>, last accessed: 12 November
506 2025) and may be granted upon signing a software license agreement with ECMWF.
507 Due to licensing restrictions, access is currently limited to European users. The TM5-
508 MP version 1.2 code with modifications for the VBS framework of this work can be
509 found at Zenodo (<https://doi.org/10.5281/zenodo.18254761>). The lite configuration of
510 the ORACLE v1.0 code, which is part of the ECHAM/MESSy Atmospheric
511 Chemistry (EMAC) model, can be obtained by applying for an EMAC license, or
512 upon request by emailing A. Tsimpidi (a.tsimpidi@fz-juelich.de). More information
513 can be found on the MESSy Consortium website (<http://www.messy-interface.org>).
514 ISORROPIA-lite is available upon request at <https://isorroopia.epfl.ch>. Post-processing
515 scripts used for creating the main figures of the manuscript are available at Zenodo
516 (<https://doi.org/10.5281/zenodo.18255310>).

517

518 **Data availability**

519 The data used for the model evaluation in Figs. 7 and 8 are from two freely available
520 observational networks: the United States Interagency Monitoring of Protected Visual
521 Environments (IMPROVE; <https://views.cira.colostate.edu/fed/QueryWizard/>, last
522 access: 2 June 2025) for fine organic mass in the IMPROVE Aerosol dataset and the
523 European Monitoring and Evaluation Project (EMEP; [https://ebas-data.nilu.no/](https://ebas-data.nilu.no/Default.aspx)
524 [Default.aspx](https://ebas-data.nilu.no/Default.aspx), last access: 2 June 2025) for PM_{2.5} organic carbon in the EMEP
525 framework. The data produced in the study are available from the authors upon
526 request.

527



528 **Competing interests**

529 The authors declare that they have no conflicts of interest.

530

531 **Author contributions**

532 SK contributed to the implementation of the ORACLE-lite module into the model,
 533 conducted the CTM simulations, analyzed the results, and wrote the paper. SM
 534 designed the study, integrated the ORACLE-lite code into the model, conducted the
 535 ESM simulations, and also contributed to writing the paper. APT and VAK provided
 536 the ORACLE-lite code, supported its integration into the model, and contributed to
 537 manuscript preparation. SNP supervised the study and contributed to manuscript
 538 writing.

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931 **Table 1:** Overview of the ORACLE-lite module characteristics, including volatility
 932 classification, evolution processes, and OA formation types for each emission
 933 class

Emissions	C^* ($\mu\text{g m}^{-3}$)	Representative volatility bin ($\mu\text{g m}^{-3}$)	Evolution in ORACLE-lite	OA type
LVOCs	10^{-2} - 10^{-1}	10^{-2}	Gas/particle partitioning	POA
SVOCs	10^0 - 10^2	10^1	Gas/particle partitioning and aging	POA and SOA
IVOCs	10^3 - 10^6	10^4	Aging and gas/particle partitioning	SOA
VOCs	$>10^6$	$>10^6$	Aging and gas/particle partitioning	SOA

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Table 2: Global budgets, atmospheric burdens and lifetimes of **(a)** POA, **(b)** SOA-sv,
(c) SOA-iv, **(d)** bSOA-v for EC-Earth during 2000-2010 and TM5-MP during
 2005 with the VBS configuration

	EC-Earth (2000-2010)	TM5-MP (2005)
LVOCs emissions (Tg yr^{-1})	7.40±3.98	7.54
SVOCs emissions (Tg yr^{-1})	31.42±10.89	31.81
IVOCs emissions (Tg yr^{-1})	52.78±9.34	53.13
(a) POA		
Evaporation (Tg yr^{-1})	3.57±0.35	3.81
Dry deposition (Tg yr^{-1})	3.48±0.79	3.26
Wet deposition (Tg yr^{-1})	31.70±8.52	31.58
Atmospheric burden (Tg)	0.73±0.04	0.69
Lifetime (days)	6.85	6.49
(b) SOA-sv		
Production (Tg yr^{-1})	19.62±1.67	19.83
Dry deposition (Tg yr^{-1})	2.02±0.17	1.97
Wet deposition (Tg yr^{-1})	17.56±2.63	17.41
Atmospheric burden (Tg)	0.60±0.03	0.60
Lifetime (days)	11.13	11.38
(c) SOA-iv		
Production (Tg yr^{-1})	38.28±7.32	37.02
Dry deposition (Tg yr^{-1})	3.90±0.57	3.37
Wet deposition (Tg yr^{-1})	34.26±6.39	32.39
Atmospheric burden (Tg)	1.35±0.07	1.21
Lifetime (days)	12.92	12.33
(d) bSOA-v		
Production (Tg yr^{-1})	51.31±1.24	52.34
Dry deposition (Tg yr^{-1})	0.69±0.07	0.40
Wet deposition (Tg yr^{-1})	50.67±6.70	50.58
Atmospheric burden (Tg)	1.15±0.06	1.18
Lifetime (days)	8.19	8.42

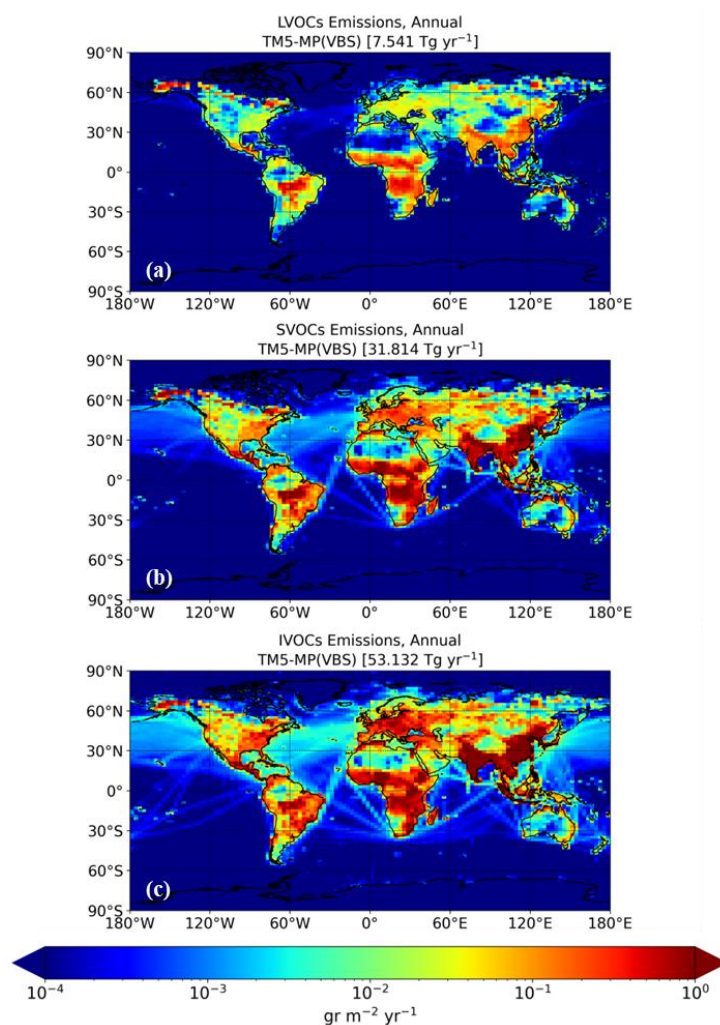
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960 **Table 3:** Evaluation metrics comparing monthly averaged predicted PM_{2.5} OA
 961 concentrations with IMPROVE and EMEP observations for the simulations of
 962 TM5-MP and EC-Earth during 2005

Simulations	Number of measurements	Mean observed ($\mu\text{g m}^{-3}$)	Mean predicted ($\mu\text{g m}^{-3}$)	MB ($\mu\text{g m}^{-3}$)	MAGE ($\mu\text{g m}^{-3}$)	NMB (%)	NME (%)	RMSE ($\mu\text{g m}^{-3}$)
TM5-MP (Default)	2124	2.11	1.54	-0.57	0.89	-27.1	42.0	1.57
TM5-MP (VBS)	2124	2.11	1.83	-0.28	0.82	-13.2	38.9	1.50
EC-Earth (Default)	2124	2.11	1.57	-0.54	0.96	-25.7	45.6	1.68
EC-Earth (VBS)	2124	2.11	1.94	-0.17	0.94	-8.1	44.5	1.61

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976 **Figure 1.** Annual emissions (in $\text{gr m}^{-2} \text{yr}^{-1}$) of: **(a)** low volatile organic compounds
 977 (LVOCs), **(b)** semi-volatile organic compounds (SVOCs), and **(c)** intermediate
 978 volatile organic compounds (IVOCs) applied in the standalone TM5-MP simulation
 979 during 2005.

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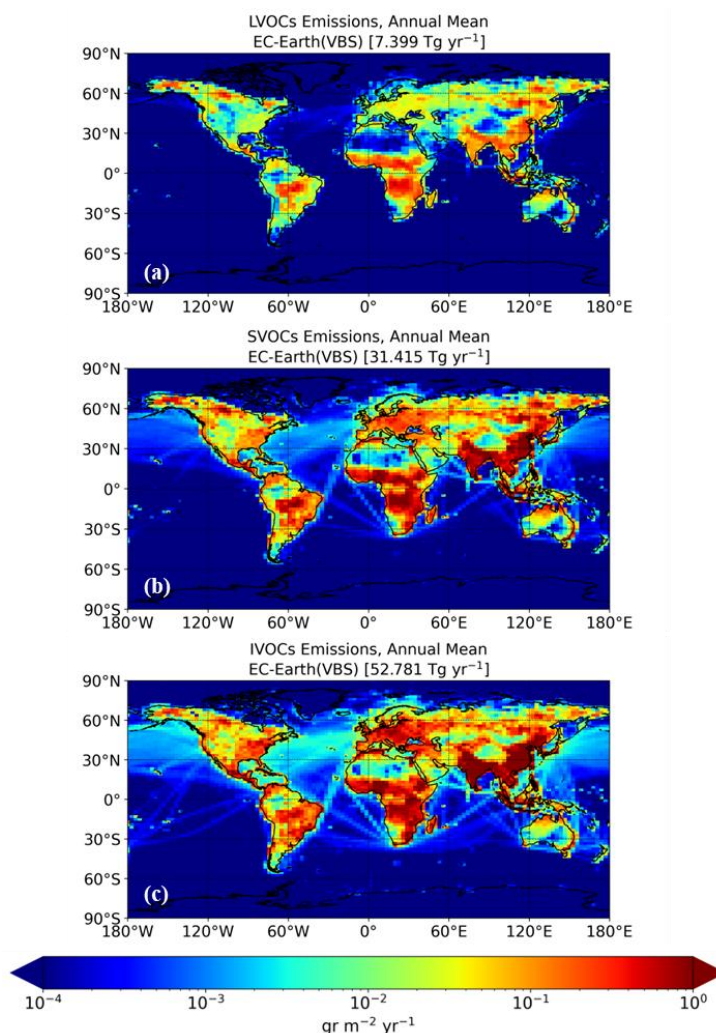
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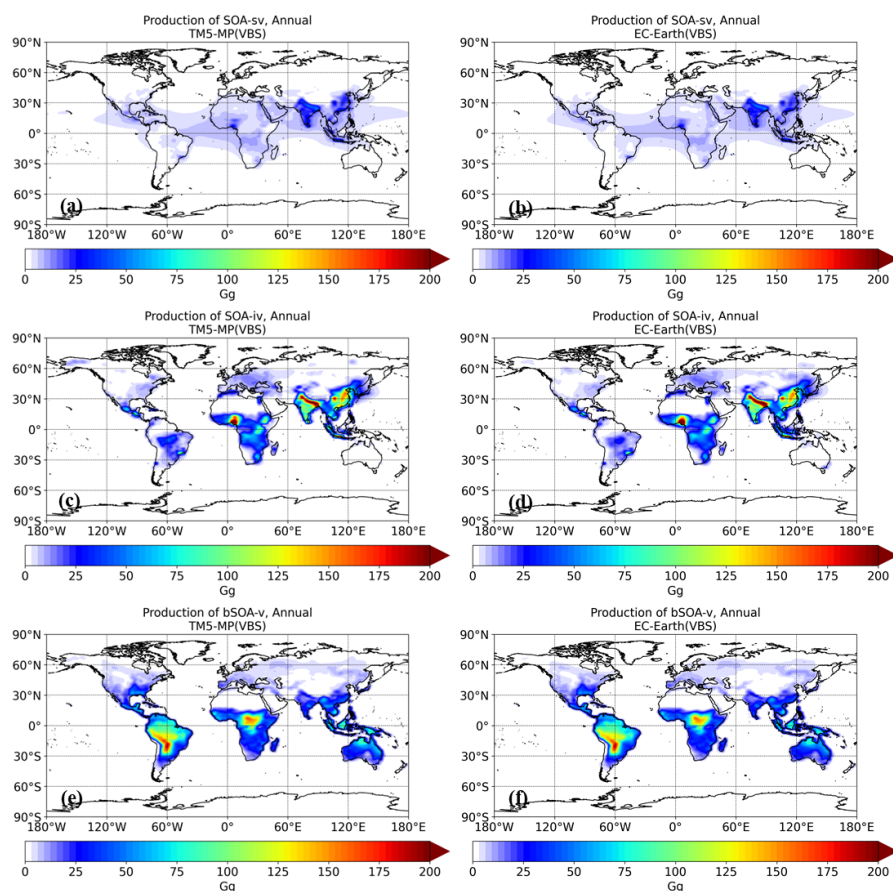
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987 **Figure 2.** Annual mean emissions (in $\text{gr m}^{-2} \text{yr}^{-1}$) of: (a) low volatile organic
 988 compounds (LVOCs), (b) semi-volatile organic compounds (SVOCs), and (c)
 989 intermediate volatile organic compounds (IVOCs) applied in the EC-Earth simulation
 990 during 2000-2010.

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993 **Figure 3.** Annual production of SOA (in Gg) as simulated using the VBS
 994 configuration of TM5-MP during 2005 (left column) and the corresponding EC-Earth
 995 predictions during 2000-2010 (right column) for: (a), (b) SOA-sv, and (c), (d) SOA-
 996 iv. For completeness, annual SOA production from biogenic VOCs (bSOA-v) in
 997 panels (e) and (f) is also shown.

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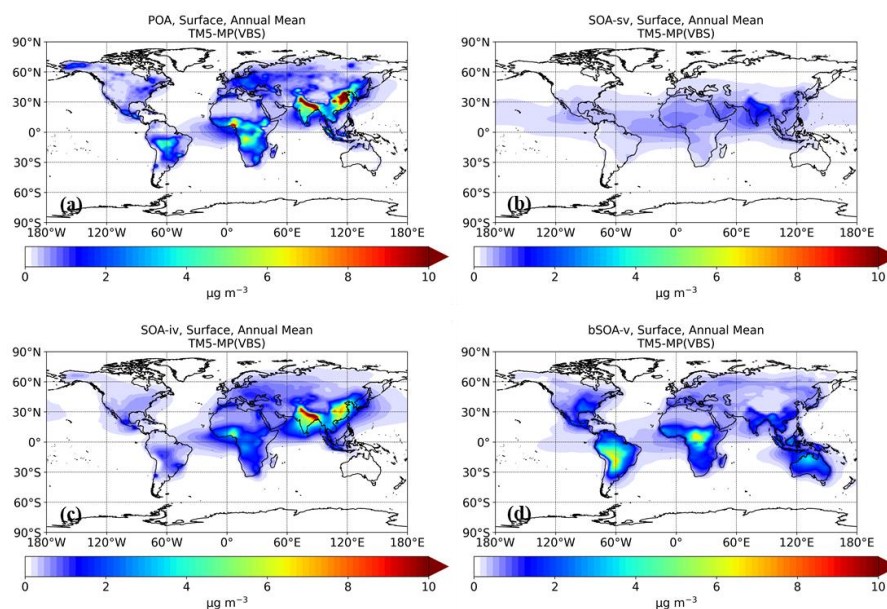
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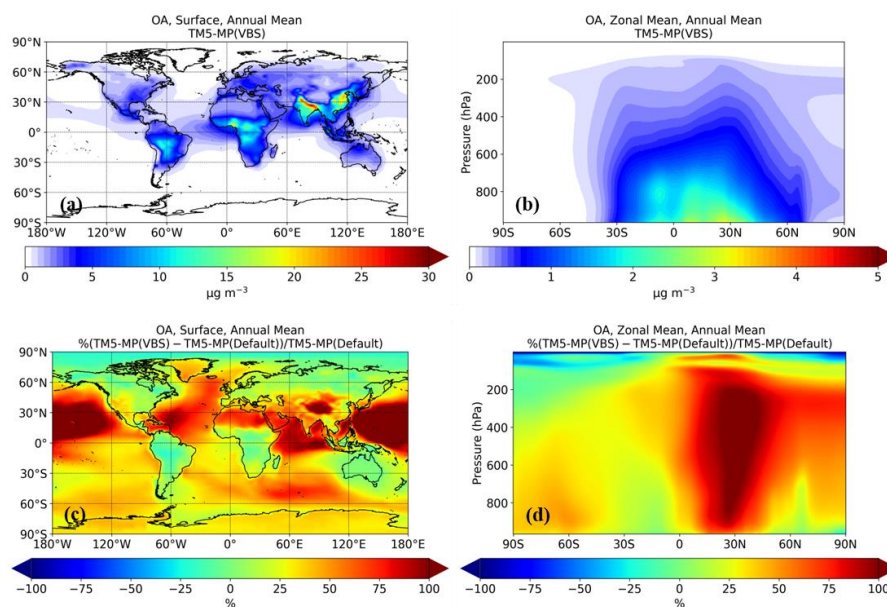


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1007 **Figure 4.** Annual mean surface concentrations (in $\mu\text{g m}^{-3}$) of: (a) POA, (b) SOA from
 1008 SVOCs (SOA-sv), and (c) SOA from IVOCs (SOA-iv) as simulated using the VBS
 1009 configuration of TM5-MP during 2005. For completeness, SOA concentrations from
 1010 biogenic VOCs (bSOA-v) in panel (d) are also shown.

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1014 **Figure 5.** Annual mean concentrations of total organic aerosol (in $\mu\text{g m}^{-3}$): (a) surface
 1015 concentrations, and (b) zonal values as simulated using the VBS configuration of
 1016 TM5-MP during 2005. Panels (c) and (d) show the corresponding relative differences
 1017 (in %) compared to the previous (default) model configuration. A positive change
 1018 indicates that the VBS configuration predicts more than the default.

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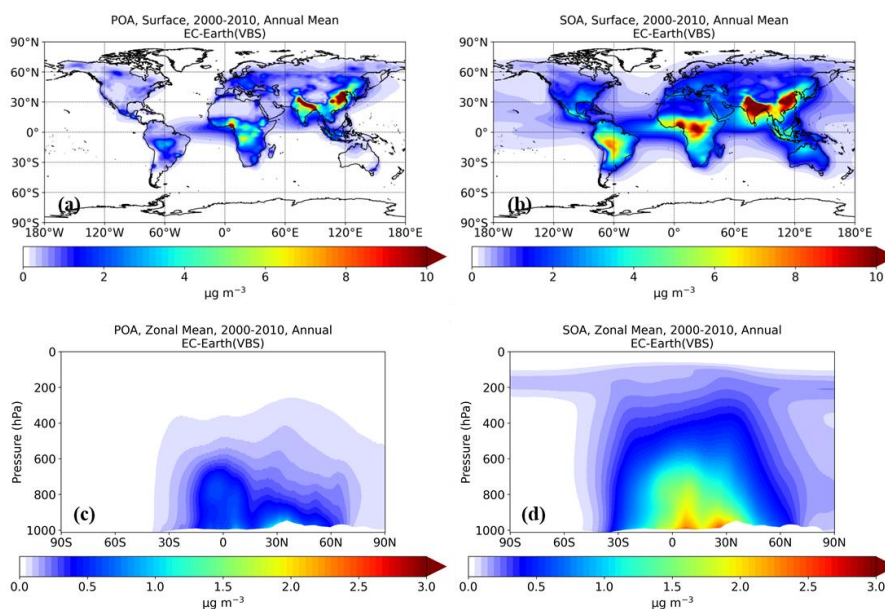
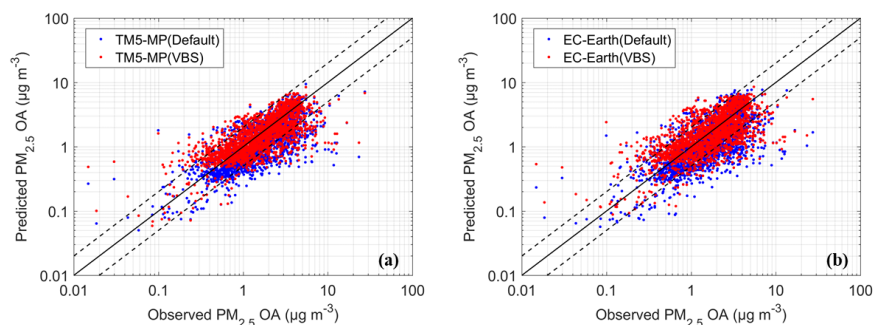


Figure 6. Annual mean concentrations of POA and SOA (in $\mu\text{g m}^{-3}$): (a), (b) surface concentrations, and (c), (d) zonal values as simulated using the VBS configuration of EC-Earth during 2000-2010.



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1052 **Figure 7.** Organic mass concentrations from simulations of: **(a)** TM5-MP, and **(b)**
 1053 EC-Earth during 2005. Scatterplots compare predicted PM_{2.5} OA concentrations (in
 1054 $\mu\text{g m}^{-3}$) with observations from the IMPROVE and EMEP monitoring networks.
 1055 Models results are shown for the default configuration (blue) and the VBS
 1056 configuration (red). Each point represents a monthly average value at a monitoring
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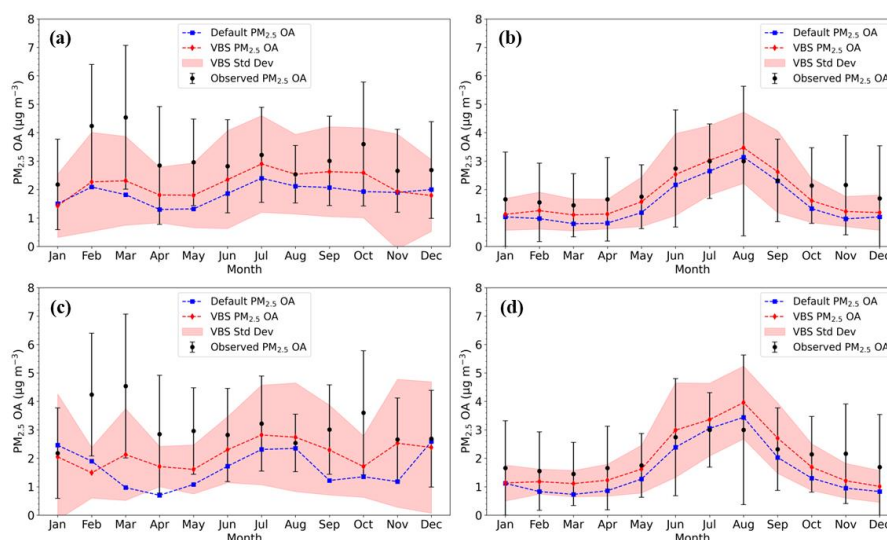
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1073 **Figure 8.** Annual cycles of monthly mean $\text{PM}_{2.5}$ OA concentrations during 2005. The
 1074 top row shows results from the standalone TM5-MP simulations at: **(a)** EMEP sites
 1075 and **(b)** IMPROVE sites, while the bottom row shows results from the EC-Earth
 1076 simulation at: **(c)** EMEP sites and **(d)** IMPROVE sites. The red line represents the
 1077 mean predicted by the VBS simulation, with red shading indicating the standard
 1078 deviation. The blue line represents the mean predicted by the default simulation.
 1079 Black dots show the observed mean values, with vertical lines showing the
 1080 corresponding standard deviations.

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