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Naturally driven variability in the global secondary organic aerosol over a decade

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Abstract

In order to investigate the variability of the secondary organic aerosol (SOA) distributions and budget and provide a measure for the robustness of the conclusions on human induced changes of SOA, a global 3-dimensional chemistry transport model describing both the gas and the particulate phase chemistry of the troposphere has been applied. The response of the global budget of SOA to temperature and moisture changes as well as to biogenic emission changes over a decade (1984–1993) has been evaluated. The considered emissions of biogenic non-methane volatile organic compounds (VOC) are driven by temperature, light and vegetation. They vary between 756 and 810 TgC y^{-1} and are therefore about 5.5 times higher than the anthropogenic VOC emissions. All secondary aerosols (sulphuric, nitrates and organics) are computed on-line together with the aerosol associated water. Over the studied decade, the computed natural variations (8%) in the chemical SOA production from biogenic VOC oxidation equal the chemical SOA production from anthropogenic VOC oxidation. This computed variability results from a 7% increase in biogenic VOC emissions combined with 8.5% and 6% increases in the wet and dry deposition of SOA and leads to about 11.5% increase in the SOA burden of biogenic origin. The present study also demonstrates the importance of the hydrological cycle in determining the built up and fate of SOA in the atmosphere. It also reveals the existence of significant positive and negative feedback mechanisms in the atmosphere responsible for the non linear relationship between emissions of biogenic VOC and SOA burden.

1. Introduction

Aerosols exert various impacts on the earth system affecting climate, atmospheric chemistry, nutrients cycling, visibility and human health. An important fraction of aerosol that is only recently considered in global model simulations is the secondary organic aerosol (SOA) (Kanakidou et al., 2000, 2004; Chung and Seinfeld, 2002; Tsi-

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garidis and Kanakidou, 2003). All global modelling studies agree that a major precursor of SOA is biogenics (Griffin et al., 1999; Kanakidou et al., 2000; Chung and Seinfeld, 2002; Tsigaridis and Kanakidou, 2003; Derwent et al., 2003; Bonn et al., 2003; Lack et al., 2004). Emissions from the terrestrial biosphere are affected by meteorological conditions like sunlight, temperature, moisture etc (Guenther et al., 1995; Naik et al., 2004). Earth's climate and atmospheric circulation are known to be subject to large natural variability that also reflects to the atmospheric composition as demonstrated by ice core measurements (Falkowski et al., 2000). This variability is expected to affect SOA levels in the atmosphere that in turn, together with the other aerosol components, impact on radiation and on the hydrological cycle (Kanakidou et al., 2004 and references therein). Thus, SOA might be involved in significant chemistry/climate feedbacks that enhance stability or perturbation of the atmosphere and climate, and that deserve careful investigation (Kulmala et al., 2004). A critical step in understanding SOA behaviour in the atmosphere is the evaluation of the importance of the human induced changes in SOA that presupposes the understanding and evaluation of the natural variability (see discussion in Falkowski et al., 2000).

The present study aims to provide the variability of the SOA distributions and thus a measure for the robustness of the conclusions on human induced changes despite the associated uncertainties in modelling SOA chemical formation in the global troposphere (Tsigaridis and Kanakidou, 2003; Pun et al., 2003; Kanakidou et al., 2004). Multiphase chemistry of organics that contributes to the SOA formation in the atmosphere (Hermann et al., 2003; Ervens et al., 2004; Tolocka et al., 2004; Kalberer et al., 2004; Claeys et al., 2004b; Barsanti and Pankow, 2004) is until now neglected in SOA global chemistry/transport model (CTM) studies due to the very limited knowledge of the relevant chemical mechanisms.

To investigate the natural component of the fluctuations of the SOA chemical production and atmospheric levels, the present study takes into account the changes (i) in meteorology as observed and (ii) in biogenic emissions. An 11-year global simulation has been performed with the TM3 model (Tsigaridis and Kanakidou, 2003) using the

European Centre for Medium-Range Weather Forecasts (ECMWF) ERA15 reanalysis meteorology for the years 1983–1993 to drive the transport, and monthly and annually varying biogenic volatile organic compounds (BVOC) emissions that have been computed by the dynamic vegetation model ORCHIDEE constrained by the ISLSCP2 (International Satellite Land-Surface Climatology Project, Initiative II Data Archive, NASA; Hall et al., 2004) satellite-based climate forcing for the corresponding years (Lathière et al., 2005[1]). All other emissions have been kept constant to those of the year 1990 (Olivier et al., 1996). Additional simulations have been performed for the years 1986 and 1990 to analyse the computed changes in the SOA budget and tropospheric burden. The choice of these two years has been based on their meteorological conditions: in general, the year 1986 has been colder and dryer than 1990, as can be seen in Fig. 1. As a consequence of the climate variability, the biogenic VOC emissions calculated by ORCHIDEE are higher in 1990 (801 TgC y^{-1}) compared to 1986 (756 TgC y^{-1}).

2. Models description

2.1. Global chemistry/transport model – TM3

The model used for the present study is the well-documented off-line chemical transport model TM3 (Houweling et al., 1998; Dentener et al., 1999; Jeuken et al., 2001). The model has a horizontal resolution of 3.75°×5° in latitude and longitude, and 19 vertical hybrid layers from the surface to 10 hPa. Roughly, 5 layers are located in the boundary layer, 8 in the free troposphere and 6 in the stratosphere. The model's input meteorology varies every 6 h and comes from the ECMWF ERA15 re-analysis data-archive (Gibson et al., 1997; http://www.ecmwf.int/research/era/ERA-15). The model has been described in detail in Tsigaridis and Kanakidou (2003), with the difference

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[1]Lathière, J., Hauglustaine, D. A., De Noblet-Ducoudré, N., Friend, A., Viovy, N., Polcher, J., and Folberth, G.: Impact of climate variability and land use changes on global biogenic volatile organic compounds emissions in a dynamic vegetation model, in preparation, 2005.

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that for the present study the spatial and temporal biogenic emissions (isoprene, terpenes and other VOC) are those calculated by the ORCHIDEE model instead of using the GEIA recommendations (Guenther et al., 1995). In addition, SOA formation from isoprene oxidation has been simulated by applying a 0.2% molar conversion factor (Claeys et al., 2004a). Multiphase chemistry that may produce additional organic aerosol mass (Kanakidou et al., 2004 and references therein) is not taken into account. This approach requires improvement, as soon as kinetic data will become available. The model includes coupled gas-phase chemistry and secondary aerosol formation calculations together with primary carbonaceous particles. Sea-salt and dust particles are not included in the model calculations.

2.2. Dynamic vegetation model – ORCHIDEE

A detailed biogenic emission scheme, based on Guenther et al. (1995) parameterizations, is integrated in the global vegetation model ORCHIDEE (Organizing Carbon and Hydrology in Dynamic EcosystEms) (Krinner et al., 2005). ORCHIDEE is composed of 3 models and is designed either to be coupled to a general circulation model or forced by climatic data. The surface-vegetation-atmosphere transfer scheme SECHIBA (Schématisation des échanges hydriques à l'interface biosphere-atmosphère), (Ducoudré et al., 1993; de Rosnay and Polcher, 1998), calculates processes characterized by short time-scales, ranging from a few minutes to hours, such as energy and water exchanges between the atmosphere and the terrestrial biosphere as well as the soil water budget. The dynamic vegetation component LPJ (Lund-Potsdam-Jena) (Sitch et al., 2003) computes long-term changes in vegetation distribution. The third component, STOMATE (Saclay-Toulouse-Orsay Model for the Analysis of Terrestrial Ecosystems), treats other processes which can be described with a time-step of a few days, such as photosynthesis, carbon allocation, litter decomposition, soil carbon dynamics, phenology, maintenance and growth respiration. Twelve plant functional types (PFTs) are considered in ORCHIDEE on top of bare soil and cohabit in each grid point depending on the geographical location. This distinction is of great

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importance since the nature and the amount of the biogenic VOC emitted are very different from one vegetation type to another.

Biogenic emissions of isoprene, monoterpenes, methanol, acetone, acetaldehyde, formaldehyde, acetic and formic acids and other volatile organic compounds (OVOC),
5   are calculated based on the well-known Guenther et al. (1995) parameterizations of which general formula is:

$$F = \text{LAI} * s * E_f * C_T * C_L * L_a \qquad (1)$$

where F is the flux of the biogenic species considered, given in μgC m^{-2} h^{-1}; LAI is the leaf area index in m^2 m^{-2}, calculated step by step by the model; unlike Guenther
10   et al. (1995) the specific leaf weight s in g m^{-2} is calculated by ORCHIDEE depending on the considered PFT; E_f is the emission factor in μgC g^{-1} h^{-1} prescribed for each PFT and biogenic species (Guenther et al., 1995, 2000; MacDonald and Fall, 1993; Kesselmeier and Staudt, 1999; Janson and de Serves, 2001). C_T and C_L are adjustment factors which account for the influence of leaf temperature and light on biogenic
15   emissions: a light and temperature dependency is considered for isoprene emissions and radiation extinction inside the canopy is taken into account, so that shaded leaves emit much less isoprene than sunlit ones, and only temperature dependency is taken into consideration for all other compounds. Thus the temperature dependency for isoprene is expressed by

$$C_T = \frac{\exp \frac{C_{T1}(T - T_S)}{R T_S T}}{1 + \exp \frac{C_{T2}(T - T_M)}{R T_S T}} \qquad (2)$$

and for all other compounds

$$C_T = \exp\left(\beta \left(T - T_S\right)\right) \qquad (3)$$

The light dependency considered only for isoprene is expressed by the factor C_L:

$$C_L = \frac{a C_{L1} Q}{\sqrt{1 + a^2 Q^2}} \qquad (4)$$

For the above mentioned equations, T is leaf temperature (K), Q is the flux of photo-synthetic active radiation (PAR; μmol phot m^{-2} s^{-1}), T_s is leaf temperature at standard conditions (303 K), R is the gas constant (8.314 J K^{-1} mol^{-1}); C_{T1} (95 000 J mol^{-1}), C_{T2}(230 000 J mol^{-1}), T_M (314 K), β (0.09 K^{-1}), α (0.0027) and C_{L1} (1.066) are empiri-cal coefficients. Since the ORCHIDEE model does not calculate the leaf temperature, we use instead the "surface" temperature, which takes into account both soil surface and vegetation energy budget.

Several studies (Guenther et al., 2000; MacDonald and Fall, 1993) underline the impact of leaf age on the emission capacity and highlight that young leaves emit less isoprene but more methanol than mature ones and that old leaves have a strongly reduced emission capacity. We thus assigned a biogenic emissions activity factor L_a (Eq. 1) for isoprene and methanol emissions, depending on the leaf age classes given in ORCHIDEE assuming that mature and old leaves emission efficiency is half the one of young leaves in the case of methanol emissions, and that young and old leaves emit 3 times less isoprene than mature ones (Guenther et al., 1999).

According to these calculations, the mean emissions over the 1983–1993 period equal 458 TgC y^{-1} for isoprene, 117 TgC y^{-1} for terpenes and 214 TgC y^{-1} for other VOC. A more detailed description and evaluation of the emission model is provided in Lathière et al. (2005)[1].

2.3. The simulations

We performed six different simulations which are summarized as follows:

1. Interannual variability. An 11 year simulation from 1983 to 1993 has been per-formed, from which the first year (1983) has been used for spin up time and the next 10 years (1984–1993) are analyzed for the present study.

2. M86/E86: This simulation corresponds to the year 1986, extracted from the 11-year simulation. Biogenic emissions of VOC and meteorological fields come from

the year 1986. This simulation has been arbitrary used as base case for the present study.

3. T90/E86: Same as M86/E86 but with temperature fields from the year 1990.

4. TH90/E86: Same as M86/E86 but with temperature and water cycle fields (relative humidity, rainfall, cloud cover) from the year 1990.

5. M90/E86: Same as M86/E86 but with meteorological fields from the year 1990.

6. M90/E90: This is the year 1990 simulation extracted from the 11-year simulation. Emissions and meteorological fields come from 1990.

SOA budgets are lower limits based on discussion in Tsigaridis and Kanakidou (2003) due to oligomers formation (Kalberer et al., 2004; Tolocka et al., 2004), multiphase chemistry and cloud processes (Warneck, 2003; Ervens et al., 2004) that are not included in the present study and are expected to enhance the SOA mass production.

3. Interannual variability in SOA budget terms for the 10-year period

The variability of the biogenic VOC emissions calculated by ORCHIDEE during the studied 10-year period (1984–1993) is shown in Fig. 2. For isoprene, monoterpenes and the other VOC categories, the minimum emissions are calculated for the year 1986 (436 TgC y^{-1}, 113 TgC y^{-1} and 207 TgC y^{-1}, respectively) and the maximum for the year 1990 (468 TgC y^{-1}, 121 TgC y^{-1} and 221 TgC y^{-1}, respectively). The increase in the emissions from 1986 to 1990 reaches 7% in all VOC categories, and is a result of the natural variability of the climate system (i.e. temperature and light intensity which are key parameters in biogenic emissions variability, and water cycle) with generally colder and dryer 1986 than 1990 (Fig. 1). These temperature and water cycle differences are not uniformly distributed around the globe; therefore the calculated emission changes vary spatially (Fig. 3).

Figure 4 depicts the annual variability of SOA budget terms as calculated by the TM3 model for the period 1984 to 1993 that results from changes in the emissions (Figs. 2 and 3) and meteorology (Fig. 1). The global annual chemical production of SOA varies similarly to the emissions of biogenic VOC and the wet deposition of aerosols (for SOA

5 is shown in Fig. 4). SOA burden in the model domain presents its minimum value of 0.143 Tg in 1986 when biogenic VOC emissions are calculated to minimize. The maximum SOA burden is however computed for the year 1991 that is subject to high BVOC emissions (although the maximum emissions are calculated for 1990) but also to somehow lower than 1990 precipitation.

10 The variation of the SOA production efficiency of the BVOC emissions for the 10-year period is shown in Fig. 5 as the ratio $\frac{\text{Global annual chemical SOAb production}}{\text{SOAb precursor VOC emissions}}$ normalized by the average ratio (0.115) for the whole period (squares). The ratio $\frac{\text{Global SOAb burden}}{\text{SOAb precursor VOC emissions}}$ normalized by the average ratio (8.26 10^{-4}) for the whole period (triangles) is also depicted as a measure of effective SOA yield of the emissions. In 1986 when the emissions are

15 the lowest of the 10-year period, the chemical production efficiency and the burden efficiency are the lowest too. An interesting remark is that for the years 1990 and 1991, when the emissions are the highest, the chemical production efficiency is below the average of the 10-year period. This could result from high removal processes during these two years, which eliminate aerosols and corresponding gas-phase species from

20 the atmosphere more efficient and thus slow down the aerosol production. The SOA production potential of the emissions is highest in the years 1987–1989 and 1992–1993. The difference of the interannual behaviour of the above discussed indices of SOA occurrence in the troposphere is largely due to the discussed involvement of the wet removal processes.

25 SOA are not only formed in the lower troposphere where most of the oxidation of the parent VOC occurs. The gas-phase products of the oxidation of the emitted biogenic or anthropogenic VOC can be transported and condense in a distant area, where the meteorological conditions favour condensation. Air circulation affects the SOA budget, since air masses are transported from areas with high temperatures to areas with

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lower temperatures, for instance towards the high troposphere, where the condensation of the semivolatile species is favoured and leads to SOA production. This process is of great importance both for the biogenic SOA (SOAb: that comes from biogenic parent VOC oxidation) and for the anthropogenic SOA (SOAa: that comes from anthropogenic parent VOC oxidation). We calculate that 60% of the net chemical production of the SOAa and 70% of the SOAb occur below about 300 hPa (first 9 model levels). This means that roughly 1/3 of the total chemical production of SOA is calculated to occur in the upper troposphere/lower stratosphere. Accordingly, corresponding SOA burden calculations show that about 60% of the SOAa and 65% of the SOAb mass is located in the lower and middle troposphere (lower layer). This vertical distribution of the SOA burden is slightly different than that of the chemical production due to enhanced removal of the particles by wet and dry deposition at the lowest altitudes. The interannual variability of these numbers is insignificant (vary about 1–2 percent units) for the whole 10-year studied period.

4. What controls the variability of the SOA budget terms

In order to study the factors that influence the interannual variability of the SOA budget terms (chemical production, destruction by deposition, total budget) five additional simulations have been performed as described in Sect. 2.3. The year 1986 (lowest emissions of biogenic VOC and chemical production of SOA for the studied years) has been chosen for comparison with 1990 (highest emissions and chemical production).

The results for the five different simulations with regard to SOA budget terms are shown in Table 1. As also indicated in this table, the chemical formation of total SOA is controlled by the biogenic VOC, since the biogenic fraction SOAb is the major contributor to the total SOA (Tsigaridis and Kanakidou, 2003). For the same amount of precursor VOC emissions (M86/E86 vs. TH90/E86) the chemical production of SOA is controlled by temperature and water cycle (relative humidity, precipitation, cloud cover), reduced by 0.4% and increased by about 2.4%, respectively, when changing from 1986

to 1990 fields. SOA production is enhanced due to reduced rainfall over most of precursor VOC source regions that results in the presence of more pre-existing particulate matter available for condensation of semivolatile compounds and thus in higher SOA mass. This points the need of accurate knowledge of the changes in the spatial pattern of the meteorological parameters and the biogenic emissions. Important feedback mechanisms are also related to transport in the atmosphere and affect SOA chemical production, which makes simulations M86/E86 and M90/E86 to have only small changes in the chemical production of SOA, i.e. transport compensates for the 2% changes calculated when comparing TH90/E86 with M86/E86.

5. SOA optical depth: spatial and temporal variability

In order to calculate the optical depth (OD) of the particles we used the approach of Kiehl and Briegleb (1993):

$$OD(\lambda) = f(RH, \lambda)B(\lambda)a \tag{5}$$

Where $OD(\lambda)$ is the OD of the aerosol at a wavelength λ, $B(\lambda)$ is the mass extinction efficiency (extinction coefficient per unit of aerosol mass at relative humidity (RH)<40%; Table 2), a is the aerosol column burden and $f(RH, \lambda)$ is the relative increase of the scattering coefficient at given RH to the scattering at low (<40%) RH (based on a polynomial fit by Veefkind (1999) for sulfate aerosols). This approach has been used for all types of aerosols that are not hydrophobic (part of carbonaceous aerosols, see Tsigaridis and Kanakidou (2003) for details). A geometric mean radius of 0.05 μm and a geometric standard deviation of 2.0 has been adopted for all aerosol types in the model.

The calculated OD of SOA (OD_SOA) presents a high spatial (Fig. 6) and temporal (not shown) variability, reflecting that of the emissions of biogenic VOC, oxidant levels and meteorological conditions as discussed in Sect. 4. The calculated differences in OD_SOA resulting from changes both in meteorology and biogenic emissions

(M90/E90 vs. M86/E86) are also shown in Fig. 6 (lower panels) and reveal high variability with both positive and negative values and an overall effect of an increased global burden of SOA.

The corresponding differences in the calculated tropospheric O_3 columns (from the surface to 300 hPa) are shown in Fig. 7 and they reflect the increased production of O_3 in VOC limited areas when VOC emissions are higher and the increased loss of O_3 where relative humidity has been increased.

Temperature is an important factor controlling the SOA distribution. The calculated differences between simulations T90/E86 and M86/E86 show the effect of temperature change between 1990 and 1986 and are depicted in Fig. 8 both for the OD_SOA and O_3 column. When temperature increases, the condensation of the semivolatile compounds present in the atmosphere is reduced, therefore the model calculates lower OD_SOA for 1990 where temperature has been increased (from 1986 to 1990; see also Fig. 1) and vice versa. Lower temperatures also imply less photochemical activity that results in reduced O_3 calculated by the model.

The atmospheric water cycle also affects the OD_SOA since it drives the wet deposition of trace gases and aerosols. Additionally, the relative humidity of the air affects the amount of water present in the aerosols, and thus their extinction coefficient. The net result of these processes is shown in Fig. 9 that depicts the spatial variation of the effect of water cycle on OD_SOA and O_3 column. In general, O_3 is increasing where the relative humidity decreases. Since O_3 reaction is the main oxidant that produces SOA in the model, an increase in the OD_SOA is also calculated.

The seasonality of the calculated OD_SOA is strong (OD_SOA during summer is more than double that of winter; Fig. 10), but the interannual variability is weak (about 12%; not shown) and of the same order of magnitude with the natural variability of the biogenic emissions. OD_SOA maximizes during summer, when both tropical and temperate/boreal forests emit large amounts of biogenic VOC, and minimizes during winter when the emissions of biogenic VOC are important only in the tropical forests that receive large amounts of solar radiation year-round. According to our model calcu-

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lations that neglect sea-salt and dust, OD_SOA contributes to the total global average OD by about 5% with other contributors being sulphate (57%), black carbon (14%), nitrate (13%) and primary organic carbon (11%). These contributions are subject to significant spatial and temporal variations.

6. Conclusions

Kanakidou et al. (2000) have shown that the human impact has drastically increased the biogenic SOA chemical production since preindustrial period. This enhanced SOAb chemical production exhibits naturally driven variations that have been studied here. This naturally driven variation in the biogenic SOA production is calculated to equal about 8% i.e. to be of the same order with the chemical production of SOA from anthropogenic VOC oxidation, whereas the SOA burden issued from biogenic VOC oxidation varies by about 11.5%.

Meteorological parameters like temperature, water cycle and mixing by transport can affect the temporal and spatial variability of SOA. In particular with regard to the chemical production of SOA from biogenic VOC oxidation, temperature affects the condensation of semivolatile compounds to the aerosol phase, while the water cycle has a major impact by affecting the removal of aerosols from the atmosphere, the water associated to the aerosol and finally the aerosol optical properties. Atmospheric circulation affects the transport of aerosols, which will change the surfaces available for condensation of the semivolatile compounds. According to our calculations this feedback mechanism seems to compensate for the effects of temperature and water cycle changes.

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Table 1. Global SOA budget and its variability between 1986 and 1990 due to meteorology and to changes in biogenic emissions.

	Burden		Wet deposition		Dry deposition		Chemical production			
	SOAa	SOAb	SOAa	SOAb	SOAa	SOAb	SOAa		SOAb	
	Gg	Gg	Tg y^{-1}	Tg y^{-1}	Tg y^{-1}	Tg y^{-1}	Tg y^{-1}	%^{a}	Tg y^{-1}	%^{a}
M86/E86	12.2	142.9	1.45	17.20	0.11	2.73	1.57	0	20.28	0
T90/E86	12.3	143.8	1.47	17.14	0.11	2.71	1.58	0.6	20.19	−0.4
TH90/E86	16.1	189.1	1.57	17.60	0.11	2.73	1.69	7.6	20.68	2.0
M90/E86	12.5	146.8	1.44	17.83	0.11	2.67	1.56	−0.6	20.26	−0.1
M90/E90	12.5	159.5	1.45	18.67	0.11	2.89	1.56	−0.6	21.94	8.2

^{a} % difference compared to M86/E86 simulation

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Table 2. Extinction coefficients at 555 nm used in the OD calculations.

Aerosol component	B (m^2 g^{-1})	Reference
SO$_4$	5	Jeuken et al. (2001)
NO$_3$	5	Jeuken et al. (2001)
BC	9	Liousse et al. (1996); Tegen et al. (1997)
Primary OA	4	Liousse et al. (1996)
SOA	4	Liousse et al. (1996)

Fig. 1. Difference in temperature (top panels), water vapour (medium panels) and precipitation (bottom panels) for the year 1990 compared to 1986, integrated over two seasons, winter (JFM, to avoid December from another year) and summer (JJA). Red means increase in the year 1990 compared to 1986 and blue means decrease.

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Biogenic VOC emissions variability

Fig. 2. Biogenic VOC emissions variability for the period 1984–1993 as calculated by the ORCHIDEE model.

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Fig. 3. Mean terpene emissions for 1986 (top panels) and differences of the mean terpene emissions (bottom panels) for winter (JFM, left panels) and summer (JJA, right panels) as calculated by the ORCHIDEE model for the year 1990 compared with those of the year 1986, in mgC m^{-2} month^{-1}.

Fig. 4. Variability of the global annual chemical production of SOA (solid squares), of the SOA burden (solid triangles), of the global annual SOA removal by wet (open squares) and dry (open triangles) deposition from 1984 to 1993.

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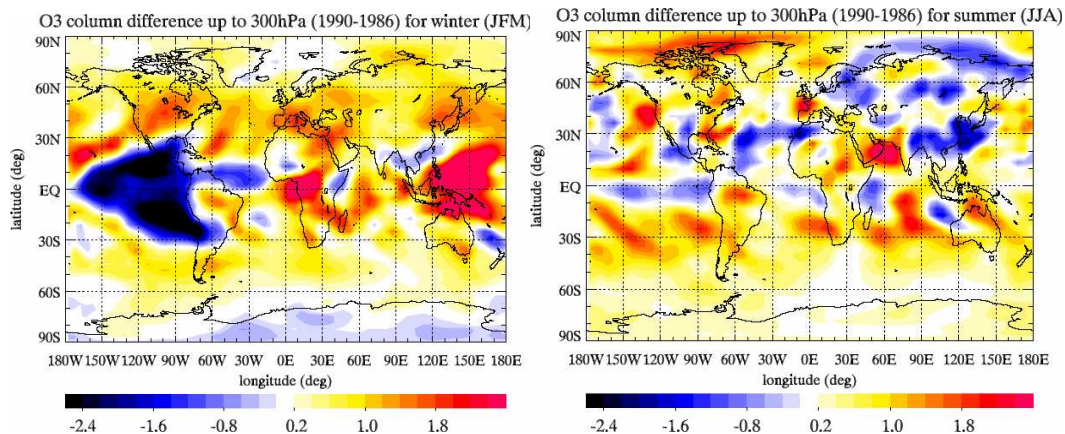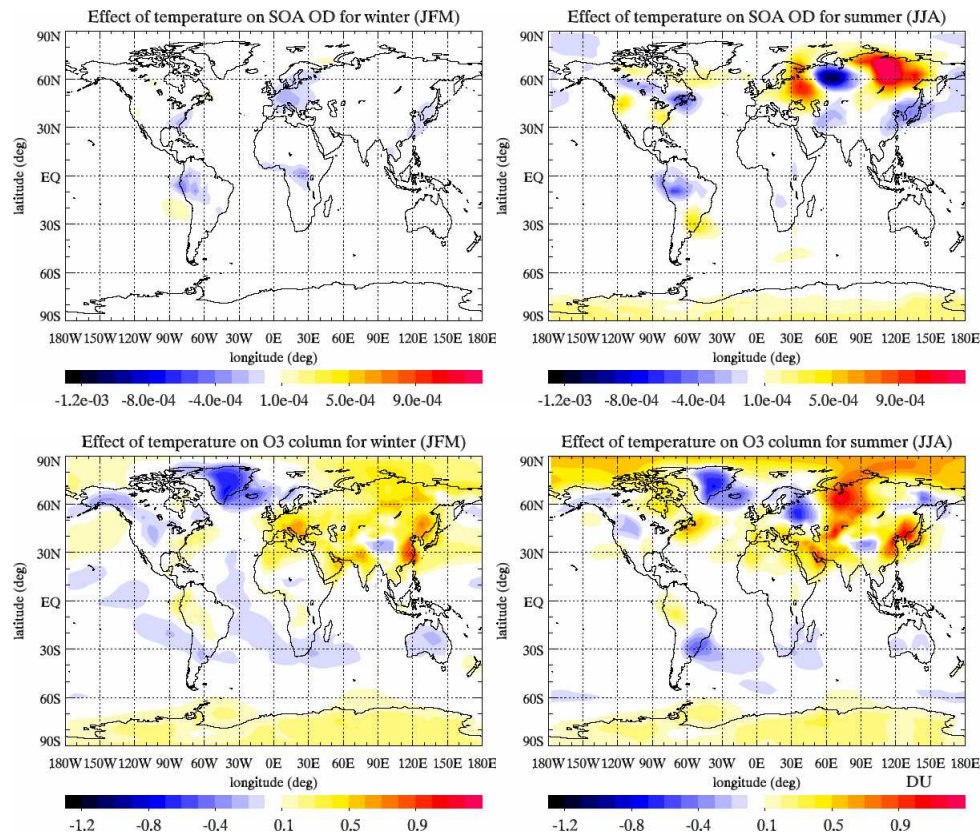**Natural variability in the global secondary organic aerosol**

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Fig. 5. Normalized ratios of the SOA chemical production (Squares) and burden (Triangles) to the VOC emissions of the specific year. The ratios are normalized dividing by the mean ratio of 8.10^{-4} and 0.115, respectively, for the period 1984–1990. Values larger (smaller) than 1 indicate higher (lower) ratios than the 10-year period mean value.

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Fig. 6. OD_SOA for 1986 (top panels) and the difference of OD_SOA for 1986 from that for 1990 (lower panels) for winter (left panels) and summer (right panels).

Fig. 7. Tropospheric O_3 column (up to 300 hPa, in DU) difference between 1986 and 1990 for winter (left panel) and summer (right panel). Red means increased O_3 in 1990 compared with 1986 and blue means reduced O_3 in 1990 compared with 1986.
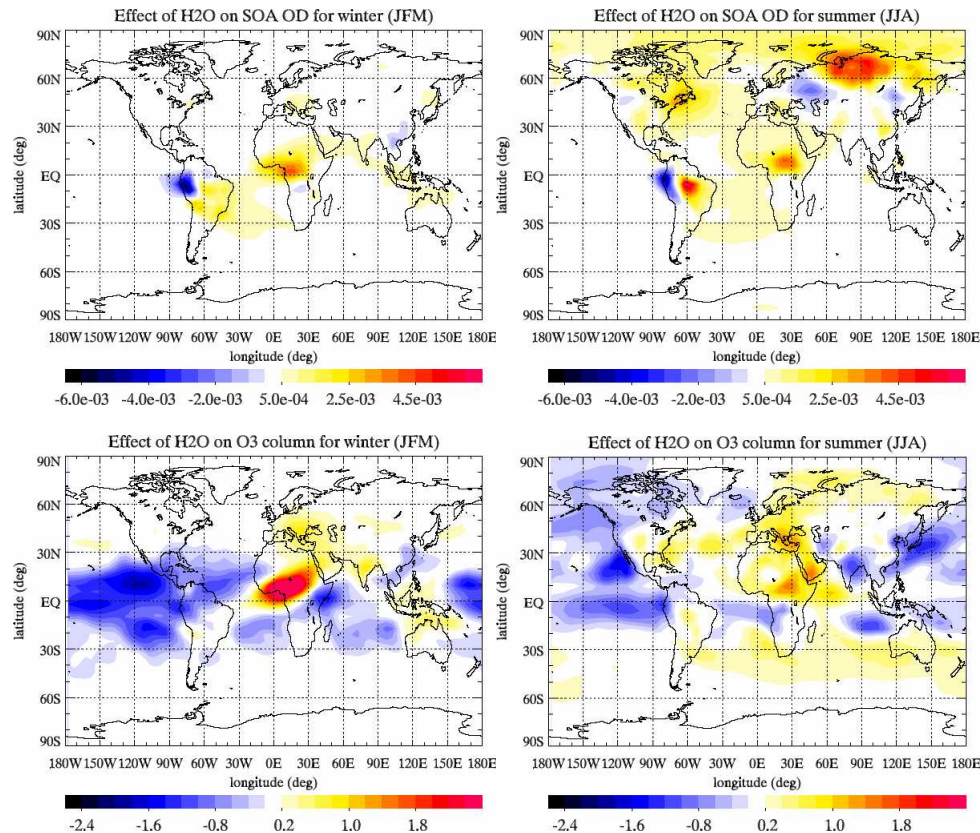
Fig. 8. Impact of temperature on OD_SOA (top panels) and O_3 (bottom panels) for winter (left panels) and summer (right panels). Red means increased OD_SOA or O_3 column resulting from the temperature of 1990 compared with that of 1986 (simulations T90/E86 vs. M86/E86, respectively). Blue means decreased OD_SOA or O_3 column resulting from the temperature of 1990 compared with that of 1986.

Fig. 9. Impact of water cycle on OD_SOA (top panels) and O$_3$ (bottom panels) for winter (left panels) and summer (right panels). Red means increased OD_SOA or O$_3$ column resulting from the water cycle of 1990 compared with that of 1986 (simulations TH90/E86 vs. T90/E86, respectively) and blue means decreased OD_SOA or O$_3$ column resulting from the water cycle of 1990 compared with that of 1986.
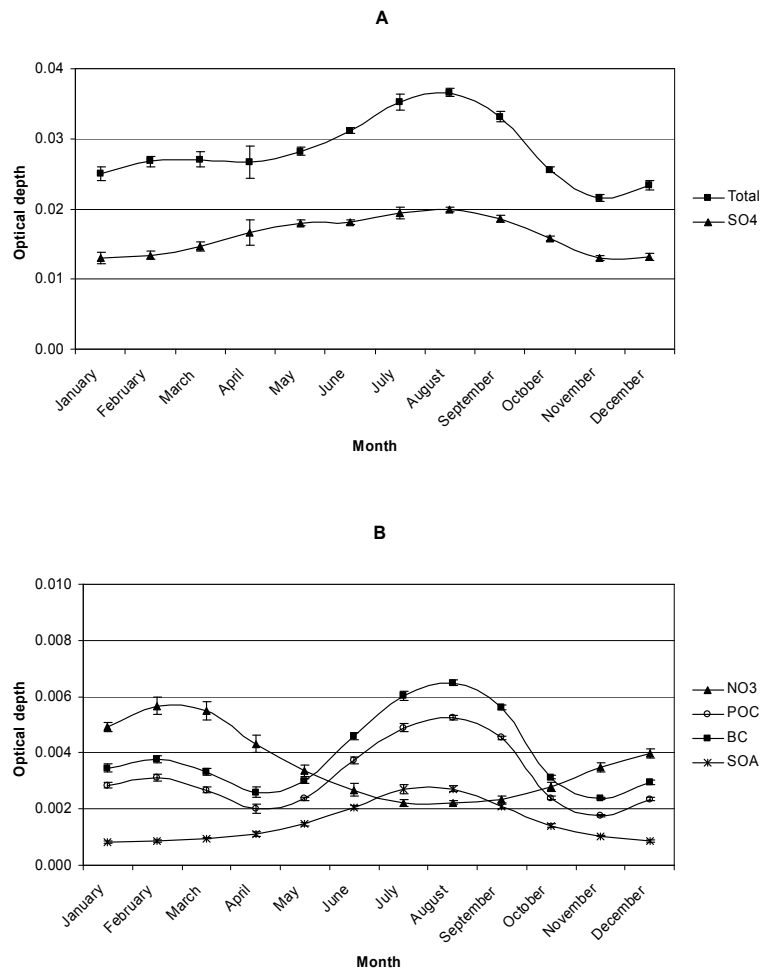
A

B

Fig. 10. Seasonal average OD for the 10-year period of 1984–1993. The error bars show the standard deviation of the interannual results. **(A)** Total and $SO_4^=$. **(B)** OD of other aerosol components.