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A simple modeling approach to study the regional impact

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A simple modeling approach to study the regional impact of a Mediterranean forest isoprene emission on anthropogenic plumes

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Abstract

Research over the past year has outlined the importance of biogenic isoprene emission in tropospheric chemistry, and notably in the context of regional ozone photo-oxidant pollution. The first part of this article deals with the development of a simple isoprene emission scheme based upon the classical Guenther's algorithm coupled with a soil-vegetation-atmosphere transfer model. The resulting emission scheme is tested in a "stand-alone" version at the canopy scale. Experimental data sets coming from Boreal, Tropical, Temperate and Mediterranean ecosystems are used to estimate the robustness of the scheme over contrasted climatic and ecological conditions. Considering the simple hypothesis used, simulated isoprene fluxes are generally consistent with field measurements and the emission scheme is thus deemed suitable for regional application. Limitations of the model are outlined as well as further improvements. In the second part of the article, the emission scheme is used on line in the broader context of a meso-scale atmospheric chemistry scheme. Dynamically idealized simulations are carried out to study the chemical interactions of pollutant plumes with realistic isoprene emissions coming from a Mediterranean oak forest. Two chemical scenarios are considered with anthropogenic emissions, respectively representative of the Marseille (urban) and Martigues (industrial) French Mediterranean areas. For the Marseille scenario, the impact of biogenic emission on ozone production is larger when the forest is situated in a sub-urban configuration (i.e. downwind distance TOWN-FOREST <30 km) and decrease quite rapidly as the distance increases. For the Martigues scenario, the biogenic impact on the plume is detectable even at a longer TOWN-FOREST distance of 100 km. For both cases, the importance of the VOC/NO_x ratio, which characterizes the aging of advected pollutant plumes over the day, is outlined. Finally, possible applications of this work for real-case studies are discussed.

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1. Introduction

Biogenic volatile organic compounds (BVOC) such as isoprene and monoterpenes are known to play key roles in atmospheric chemistry processes, ranging from local scale to global scale (Guenther et al., 1995). In particular, the tropospheric production of ozone due to photochemical reactions of nitrogen oxides (NO_x) is enhanced by the presence of BVOCs, which are very reactive compared to most of the anthropogenic VOC (Stockwell et al., 1997). The impact of biogenic emission on ozone pollution is likely to be detectable in urban area (Chameides et al., 1988) and very important in rural sites (Tsigaridis and Kanakidou, 2002). Therefore, control policies on primary pollutant emission must account for biogenic emissions as much as possible (Roselle et al., 1994). At regional scale, spatial extent and dissemination of BVOCs throughout the landscape favors complex interactions with pollutant plumes determining the regional ozone budget (Sillman et al., 1999). One important aspect of such studies is the representation of biologically regulated emission in simple models (Guenther et al., 1993) designed to simulate emissions for a wide range of vegetation and species. Another important aspect is the modeling of the diversity and the complexity of biogenic source distributions through the landscape.

The French meso-scale chemical model Meso-NH-C (Tulet et al., 1999, 2002; Crassier et al., 2000) was developed in the last few years, and has been frequently used for regional pollution studies. The present paper aims at presenting and testing a simple biogenic emission scheme, and performing Meso-NH-C simulations. Sections 2 and 3 of this work present a “stand alone” approach where isoprene emission fluxes at the canopy scale are simulated using simple parameterizations and hypothesis. The resulting BVOCCEM scheme is compared to field measurements of fluxes coming from boreal, temperate, tropical and Mediterranean isoprene emitting forest ecosystems. The objective of the two sections is to test the strength of the scheme for contrasted conditions, rather than to perform a detailed validation study for each of the different data sets.

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In the second part, the BVOCEM emission scheme is used to simulate isoprene fluxes in the broader context of the Meso-NH-C atmospheric chemistry model. The study of the potential impact of isoprene emissions from a Mediterranean oak forest on anthropogenic polluted plumes is presented using idealized applications. The anthropogenic sources of primary compounds correspond to the French Mediterranean urban areas of Marseille and Martigues. They are characterized by quite different VOC/NO_x emission ratios. The influence of distance between town and forest on ozone production is discussed for these two chemical scenarios. These anthropogenic source locations are situated in the region where the ESCOMPTE regional experiment (Cros et al., 2003) was carried out, a program designed to improve forcing and validation of regional chemical models.

2. Isoprene emission modeling

Numerous studies deal with the modeling of isoprene fluxes at the ecosystem level, using sometimes very detailed representation of canopy structure, micro-climate and chemical processes (see for example Baldocchi et al., 1999). In the present study, the isoprene emission scheme is designed for regional atmospheric chemical modeling. This implies a compromise between the realism of modeling, and the necessary restriction in the number of parameters to be used. We adopt here a classic bottom-up approach to derive isoprene fluxes at the canopy level from species emissions factors (Guenther et al., 1993, 1995, 1999). The ecosystem canopy is assimilated to a simple homogenous layer of leaves made up of the different species. With regard to such a hypothesis, and using Guenther approach, the instantaneous isoprene canopy flux is related to leaf level emissions and estimated as:

$$F(t) = EP \times EC(t), \quad (1)$$

where F is the instantaneous isoprene emission flux at the canopy scale (in $\mu\text{g m}^{-2} \text{h}^{-1}$). EP is the canopy emission potential as defined in Solmon et al. (2004).

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$EC(t)$ is the canopy environmental correction factor accounting for effect of microclimate (photosynthetically active radiation (PAR) and leaf temperature) on isoprene emissions (Guenther et al., 1993). For a given vegetation cover, the in-situ species composition, as well as the estimation of species leaf biomass are required to determine the canopy emission potential. When not available from in-situ leaf-level measurements, standardized species emission factors can be derived from existing inventories, see for example recent reviews of Owen et al. (2001), and Solmon et al. (2004) on the ESQUIF program. Numerous studies outline the large inter and intra-specific variability of species emission factor, as well as their phenologic and seasonal dependence (Xiao-shan et al., 2000; Otter et al., 2002). One must keep in mind that such uncertainties may directly affect the emission potential estimations (Guenther et al., 2000). Once determined, EP is here assumed to be constant for the time scale considered in this study (typically diurnal evolution over few day simulations).

Calculation of the above-mentioned $EC(t)$ factor is done using the vegetation atmosphere (SVAT) transfer model ISBA-Ags. The extension “Ags” (A for carbon assimilation and gs for stomatal conductance) allows to take into account an interactive vegetation for local adaptations (Calvet et al., 1998). ISBA-Ags requires a limited number of surface parameters (Noilhan and Planton, 1989) and can run in “stand-alone” simulations, or interactively in the context of the Meso-NH meso-scale atmospheric scheme (Crassier et al., 2000; Tulet et al., 1999, 2003). Based on a big-leaf approach, ISBA-Ags treats one single surface energy budget. If the cover is dense enough (e.g. a close forest), the prognostic surface temperature is assumed to represent the mean vegetation layer temperature. To perform the calculation of EC , the radiation attenuation is taken into account in the simple layer canopy. Following Roujean et al. (1996), direct and diffuse radiation influence is taken into account. Radiation at the top of the canopy, leaf area index (LAI), solar zenithal angle, leaf radiative properties and a spherical distribution of leave directions are the main parameters considered in the model. Integration of the PAR correction coefficient through the canopy is then resolved with a 3 points Gauss method (Calvet et al., 1998). Evolving isoprene fluxes are calculated for

the considered ecosystem, accounting for all these considerations.

In the following sections, the above described isoprene emission scheme is referenced as the BVOCEM scheme.

3. Stand alone validation

5 3.1. Site descriptions and forcing data

The BVOCEM emission scheme was tested initially in “stand alone” mode using different experimental datasets. Four sites representing contrasting forest ecosystems (in terms of climate and ecology) have been considered. Their main characteristics are described in Table 1. For these four different databases, temporal series of measured meteorological forcing, as well as in-situ estimations of soil water content (root zone) were used to constrain ISBA-Ags. Surface parameters (vegetation cover rate, *LAI*, height, roughness, albedo...) were determined from in-situ estimations, or derived from available primary parameters. Emission potentials were determined using the Solmon et al. (2004) methodology. References about the experimental sites are also given in Table 1, except for the last Mediterranean experiment (MED case). This site was monitored using instruments deployed for the ESCOMPTE 2001 (Cros et al., 2003) regional experiment. Specific attention was paid to the estimation of reported *LAI* and emitting leaf biomass. Several field campaigns, for example those conducted during the BEMA project, have shown that the emission potential for *Quercus pubescens* is extremely variable (e.g. Kesselmeier et al., 1999). Thus, it was necessary to estimate a representative value of *EF* during the MED experiment. No “cuvette” measurements were performed at the plant level on this ecosystem. However, the measured canopy level fluxes were normalized to standard conditions from the measured *PAR*, air temperature and emitting biomass resulting in an emission factor of about $20 \mu\text{g g}_{dw}^{-1} \text{h}^{-1}$. Of course this estimation does not constitute a direct measurement of emission factor, but it allowed the determination of its probable value which is consistent with the range

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of values found in the literature (from 22,75 up to 90 $\mu\text{g g}_{dw}^{-1} \text{h}^{-1}$). Owen et al. (1998) determined an emission factor 22,75 $\mu\text{g g}_{dw}^{-1} \text{h}^{-1}$ for *Quercus pubescens* in a French forest during the BEMA experiment from June to September 1995, (conditions very similar to the MED experiment). This latter value, which approaches the ecosystem estimation, has been considered to get the representative emission potential of 5185 $\mu\text{g m}^{-2} \text{h}^{-1}$ reported in Table 1.

3.2. Results

A key step of the validation procedure was to characterize as well as possible the canopy microclimates, especially leaf temperatures. Following Calvet et al. (1998), the mesophyllian conductance was calibrated using time series of net radiations, latent and sensible heat fluxes measurements, by minimization the root mean square between model and measurements. For the four different databases, simulated latent and heat fluxes agreed reasonably with the measurements, with a total root mean square error varying from 30 W m^{-2} for the BOR case to 70 W m^{-2} for the MED case. For the MED situation, the simulation of latent heat flux was difficult due to strong controls of Mediterranean plants on transpiration, as well as soil moisture heterogeneity. After this numerical adjustment, we assume that calculated surface temperatures through ISBA-Ags energy budget reasonably approaches mean leaf temperatures.

Figure 1 displays a time series of modeled and experimental values of isoprene fluxes for the different cases. The temporal evolution of fluxes is in quite good agreement with measurements, morning activation, diurnal maxima and evening decrease generally in phase. However, the model tends to systematically overestimate the measurements (Fig. 2). Since the ecosystem has a homogeneous species composition, and we are confident with the estimation of the emission potential as described in Sect. 3.1 (see Table 2 for correlation coefficients), we assume that the error is mainly due to the canopy environmental correction factor calculation. Particularly, the assumption of a single leaf energy balance does not account for “shade leaves” in the canopy,

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which are cooler than sun exposed leaves. The errors caused by this is enhanced by the fact that leaf level emission rate varies through the canopy in function of leaf age and phenology. The results can be improved by assuming that leaf temperature is equal to air temperature measured at the same level (which was checked in situ, data not shown).

The scatter plot in Fig. 2 presents the results obtained on the four studied ecosystems. It outlines the tendency for overestimation of simulated fluxes as described above. However, with regards to the numerous uncertainties of biogenic emissions, simulated isoprene fluxes remains acceptable for the four ecosystems.

In the following section, the use of the simple BVOCEM model in the context of the meso-scale MESO-NH-C atmospheric chemistry model is described, with a specific focus on biogenic impact on ozone formation.

4. Application: chemical impact of a Mediterranean forest isoprene emission

4.1. The Meso-NH-C model

The non-hydrostatic meso-scale meteorological model Meso-NH was jointly developed by CNRM (Meteo France) and Laboratoire d'Aérodologie (CNRS) (Lafore et al., 1998). The soil surface scheme used in the model is the above-mentioned ISBA model, which can be activated with the A_{gs} option (interactive vegetation). The BVOCEM model described in Sect. 2 has been implemented in the Meso-NH framework to derive biogenic emissions. These on-line surface emissions complement the anthropogenic fluxes (from GENEMIS database) included into the chemical scheme of Meso-NH, summarized in Shure et al. (1998) and Tulet et al. (1999). Chemical mechanisms are described by the Regional Lumped Atmospheric Chemical Scheme (ReLACS) derived from the Regional Atmospheric Chemistry Mechanism (RACM) model (Stockwell et al., 1998). ReLACS includes 37 species (biogenic hydrocarbons being explicitly differentiated) associated with 128 chemical reactions (Crassier et al., 2000; Tulet et al., 2003). On-line

coupling with Meso-NH creates Meso-NH-Chemistry (Meso-NHC) to perform meso-scale simulations.

4.2. The configuration of idealized simulation

The main objective of the following simulations is to characterize the impact of biogenic emissions in a simplified dynamical situation with, as far as possible, realistic surface chemical emissions for a Mediterranean region. We considered a flat idealized domain of 250 km², with a 5 km horizontal resolution and a 30-levels vertical grid with a fine stretched resolution starting from 10 m above the surface. The top of the domain is situated at 10 500 m. Three types of surface were defined on this domain (Fig. 3): (i) the TOWN type associated with the town energy budget model of Masson et al. (2000), (ii) a default vegetation associated with a set of surface parameters representative of Mediterranean grassland (Masson et al., 2000), and (iii) a FOREST cover characterized by the set of surface parameters described in the above-mentioned MED experiment (see the stand alone study in Sect. 3). The dimensions of the TOWN and the FOREST patches are typical of landscape configuration possibly found in European regions (here, the ESCOMPTE region). The atmospheric variables (temperature, humidity and pressure) are initialized with the ECMWF analysis and corresponds to 30 July 2001. Boundaries conditions of the domain are constrained every 6 h using the ECMWF analysis. The wind components were set to 3 m s⁻¹, in order to simulate a light breeze, advecting primary anthropogenic pollutants to the forest region. The simulations were calculated for the period 30 July, 00:00 UT to 31 July, 00:00 UT.

4.3. Chemical sources and initial conditions

For TOWN anthropogenic emissions, we considered successively two distinct sources, both provided by the GENEMIS primary compounds database (Wickert et al., 1999) and representative of a typical summer day. The first corresponds to the Marseille town (referred as *MS* scenario in the following). The *MS* NO_x and VOC emissions are

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represented in Figs. 4a and 4b. The VOC term represents the sum of primary organic groups as defined in Crassier et al. (2000). The corresponding VOC/NO_x emission ratio is about 5 ppbC/ppb during the maximum diurnal emission (Fig. 4c). The second source is related to the Martigues area (referred as *MT* scenario). It corresponds to an industrialized zone, associated with significant car traffic. The *MT* emissions do not show a strong diurnal cycle when compared to *MS* urban emissions (Figs. 4a and 4b). The main difference between these anthropogenic sources is the VOC/NO_x ratio which is about 2 for the *MT* and about 4 for the *MS* scenario (Fig. 4c). During the simulations these anthropogenic emissions are prescribed hourly at the surface on the TOWN delimited area.

As for biogenic emissions, the diurnal FOREST isoprene emission is directly calculated by the previously-described BVOCEM scheme. Figure 5 shows that the on-line simulated fluxes are in good agreement with measured fluxes coming from the MED experiment (see Sect. 3), thus providing a realistic biogenic source of isoprene for the considered typical summer day.

The initialization of chemical fields was based on clean air values for each species of the ReLACS scheme. In particular, the initial homogeneous ozone concentration was set to 40 ppb all over the domain. The same clean air characteristics are considered for the boundary conditions. Consequently, the upwind ozone concentrations reaching the TOWN are constant, and equal to 40 ppb.

In order to study the scale of interactions between anthropogenic plumes and biogenic sources, two sets of simulation were performed for the *MS* and *MT* anthropogenic sources. Four different downwind distances (referred as d_1 -the shortest, d_2 , d_3 and d_4 -the longest) between the FOREST and TOWN area were considered (Fig. 3). For each distance, simulations with biogenic isoprene emission (referred as *BIO* simulations) and simulations without biogenic emission (*NOBIO* simulations) were calculated, chemical and dynamical conditions being strictly equivalent.

4.4. Reference anthropogenic plumes

The *NOBIO* simulations performed on *MS* and *MT* scenarios show the development of anthropogenic ozone plumes. In Figs. 6 and 7, these plumes are characterized by surface ozone concentration exceeding the initial clean air value of $[O_3]=40$ ppb.

5 The *MS-NOBIO* ozone plume shows a “typical” development over the day with a concentration increase starting around 09:00 UT, and a maximum concentration of 60 ppb reached around 12:00 UT (Fig. 6a). The maximum ozone concentration is characterized by with a NO_x concentration about 2.5 ppb and VOC/NO_x ratio around 6.5 (Figs. 6b and 6c). Then, the NO_x plume extension (Fig. 6b 12:00 to 15:00 UT)
10 reduces as the ozone plume extends in the afternoon, higher NO_x concentrations are only found near the anthropogenic TOWN source.

The ozone concentrations in the *MT-NOBIO* scenario are lower than in the *MS-NOBIO* scenario. However, the area covered by the *MT-NOBIO* ozone plume is larger than the *MS-NOBIO* plume for all times of the day. Until 12:00 UT, the ozone concentration increase is mainly observed at the front and on the edge of the anthropogenic plume (Fig. 7a). The “ozone gap” inside the plume during this time is accompanied by high NO_x concentrations (>8 ppb) and $VOC/NO_x < 2.5$ (Figs. 7b and 7c). Comparing with the *MS-NOBIO* simulations, the maximum ozone concentration is reached at 15:00 UT and for a farther downwind distance. As dynamical conditions are equivalent, this must be related to the larger nighttime anthropogenic precursor fluxes in *MT*
15 scenario as in *MS* scenario (Figs. 4a and 4b).

20 In the following discussion, *NOBIO* plumes will be considered as reference to evaluate the impact of isoprene emissions in *BIO* simulations.

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4.5. Biogenic impact on the anthropogenic ozone plume

4.5.1. Ozone concentrations

Figures 8a and 8b represents the diurnal cycle of ozone maximal concentration in the domain obtained when taking into account isoprene emission (*BIO* simulations), for different TOWN-FOREST distances (d_1 – the shortest to d_4 – the longest) and for *MS* and *MT* scenarios. The relative contribution of isoprene emission to ozone maximal concentration cycle is calculated with

$$W_c = \frac{[O_3]_{\max}^{BIO} - [O_3]_{\max}^{NOBIO}}{[O_3]_{\max}^{BIO}}.$$

The evolution of W_c over the daytime is shown in Figs. 8c and 8d.

MS scenario

The larger impact of biogenic emission in terms of ozone concentration is obtained for the distance d_1 with a diurnal increase of W_c reaching 28% to 30% between 11:00 and 12:00 UT (Fig. 8a). A VOC/NO_x ratio <2.5 with significant NO_x concentrations, represented in Figs. 6b and 6c (*MS-NOBIO* simulation), characterizes a “VOC sensitive” chemical regime in the plume (Sillman, 1999). If isoprene emissions (*MS – BIO* simulation) are taken into account, the “VOC sensitive” regime encourages high ozone formation. This impact is weaker for the d_2 distance ($W_c=8$ to 9% in Fig. 8a) since the reference NO_x concentration and the VOC/NO_x ratio increases rapidly with downwind distance (Figs. 6b and 6c). After 12:00 UT, the reference VOC/NO_x ratio characterizing the anthropogenic plume at d_1 and d_2 increases with time (Fig. 6c) as NO_x concentrations decrease (Fig. 6b). W_c decreases consequently in spite of higher isoprene emissions in the afternoon. For the distances d_3 and d_4 , the reference pollutant plume has already evolved towards a “NO_x sensitive” regime

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(Sillman, 1999) when arriving at the FOREST area. Thus, there is nearly no impact of isoprene emissions on the ozone concentration (Fig. 8a).

Figure 9a shows the ozone concentration field when taking into account the biogenic emissions in the d_1 configuration. The effect of isoprene emissions on the shape and the extension of the ozone plume appears evident from the comparison between Figs. 6a and 9a. This effect is more pronounced in *BIO* simulations, and the position of the maximum ozone concentration is also shifted downwind. When biogenic emissions are taken into account, the ozone maximal concentration in the afternoon may be related to the moderate dilution of the ozone plume (Fig. 8a). Figure 9b illustrates the diurnal cycle of the VOC/NO_x ratio when biogenic emissions are considered. The comparison between Figs. 6c and 9b shows that the interaction with the FOREST area tends to accelerate the increase of the VOC/NO_x ratio, and thus, the maturation of the pollutant plume from a “VOC sensitive” regime towards a “NO_x sensitive” regime, which is characteristic for “rural pollution” (Sillman, 1999).

MT scenario

The reference *MT-NOBIO* simulation at 09:00 UT is characterized by a VOC/NO_x ratio <2.5 (Fig. 7c). This latter condition defines a VOC sensitive regime, especially for the d_1 , d_2 and d_3 distances and to a lesser extent for d_4 (Fig. 8a). Consequently, in the *MT - BIO* simulations, the ozone formation is sensitive to even small amounts of isoprene emissions (Fig. 8d). However, in the *MT-NOBIO* NO_x saturated plume, the ozone production is limited by the high NO_x concentrations (Sillman, 1999) as the plume approaches the TOWN (Fig. 7b). Thus, the maximum biogenic impact in the morning, in the *MT-BIO* simulation, is obtained for the distance d_3 , with W_c reaching 20% at 11:00 UT (Fig. 8d). When anthropogenic and biogenic sources are separated by the distance d_3 , the reference NO_x concentrations decrease sufficiently to reach a VOC/NO_x ratio favorable to ozone formation (Figs. 7b and 7d). Maximum W_c and maximum ozone concentration, calculated in parallel for the four distances,

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will occur during the day first for the distance d_3 followed by d_2 and d_1 (Figs. 8b and 8d). The calculation depends on the plume degree of maturation with VOC/NO_x ratios characteristic of high ozone concentrations. For the distance d_1 (i.e. very close to the TOWN source), the anthropogenic plume is never NO_x limited, and the maximum W_c of 25% is obtained around 14:00 UT, i.e. when the isoprene emission is at the maximum. The highest value of the maximum ozone concentration is also obtained for the d_1 distance between 15:00 UT and 17:00 UT (Fig. 8b). As illustrated on Figs. 7a and 10a, the shape of the ozone plume is significantly modified by the biogenic source: the ozone gap tends to be reduced by the adjunction of biogenic VOCs and the maximum concentration location is modified. As discussed for the *MS* scenario, diurnal VOC/NO_x ratio cycle is also strongly affected, indicating an acceleration of the downwind plume maturation due to isoprene emissions (Fig. 7c vs. Fig. 10b).

4.5.2. Integrated ozone amount

In Sect. 4.5.1, the impact of biogenic sources is discussed in term of concentrations, which are relevant for the comparison with pollution thresholds. However, maximum ozone concentrations translate a localized impact in space (in our study, at the proximity of FOREST) and time (when the chemical regime is optimal). The evaluation of total ozone contained and produced in the plume Q_{O_3} (kg) can yield a more integrative information about the ozone formation. Q_{O_3} (kg) has been calculated for the whole 3-dimensional domain as the sum of ozone amounts contained in the cells for which the ozone concentration exceeds 40 ppb (i.e. the initial concentration). The relative contribution of ozone from biogenic precursors to the total amount is calculated as:

$$W_Q = \frac{Q_{O_3}^{BIO} - Q_{O_3}^{NOBIO}}{Q_{O_3}^{BIO}}.$$

MS scenario

The maximum ozone production in the plume is obtained for the d_1 configuration with a total ozone amount reaching 260.10^3 kg at 18:00 UT (Fig. 11a). The final ozone amount is about 240.10^3 kg in d_2 configuration (Fig. 11a) for the same time. The proximity of FOREST and TOWN areas favors the availability of anthropogenic NO_x reacting with biogenic compounds, as discussed in Sect. 4.5.1. Moreover, the proximity of FOREST and TOWN areas also causes a longer interaction between the anthropogenic plume and the forest area during daytime. No significant differences were observed between the configurations d_3 and d_4 cases which both result in an ozone amount of around 230.10^3 kg.

The large relative impact of biogenic emissions from 08:00 UT to 10:00 UT appears evident from the diurnal W_Q cycle (Fig. 11c). The W_Q cycle shows the sensitivity of biogenic precursors for NO_x that are available on the morning. However, the corresponding absolute ozone production is small (Fig. 11a). From 10:00 UT to 20:00 UT, the ozone production increases and the calculated W_Q are stable, around 10% for d_1 and 5% for d_2 . The biogenic contributions for the d_3 and d_4 scenarios are insignificant, as discussed in Sect. 4.5.1, no interaction between the forest emission and the anthropogenic plume for these distances.

MT scenario

In the *MT* simulation, Q_{O_3} is more important than the ozone amounts produced with the *MS* scenario. The maximum production is found for the d_1 distance, with a value reaching 350.10^3 kg at the end of the simulation (Fig. 11b). The amount of ozone in the d_2 configuration reaches 350.10^3 kg and is slightly higher compared to the d_3 case (Fig. 11b). As in the *MS* scenario, a high value of W_Q is obtained from 08:00 UT to 10:00 UT. The significant amounts of ozone produced with high value of W_Q in Fig. 11c compared to Fig. 11d, for the period 10:00 UT–14:00 UT, shows clearly

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an impact of biogenic emissions on ozone production for all distances. As outlined before, the global decrease of W_Q relies upon the VOC/NO_x ratio, which increases over the day as the ozone plume develops. The maximum biogenic impact on the ozone production is found for the d_1 distance, with a stable value of $W_Q=15\%$ (at 18:00 UT) (Fig. 11d). For the distances d_2 and d_3 , we obtain a final impact around 10% and 4% for the distance d_4 .

5. Conclusions

The first part of the paper presents a model designed to simulate isoprene fluxes at the scale of the ecosystem canopy. This scheme is based on Guenther's formulation (Guenther et al., 1993). The driving microclimatic variables are calculated using the ISBA-Ags SVAT model. This procedure results in a simple isoprene emission module and provides consistency with regional modeling approaches. Validations carried out at the scale of different ecosystem canopies (Boreal, Tropical, Temperate and Mediterranean forests) have shown a reasonable agreement between model and measurements. These results encouraged us to consider this emission scheme suitable for regional chemistry modeling. Developments for calculating specific leaf energy balance consistent with the ISBA-Ags surface energy budget are carried out on the basis of measurements collected during the ESCOMPTE 2001 field campaign.

The second part of the article deals with the use of the above defined isoprene emission scheme coupled with the atmospheric chemistry model Meso-NH-C. The goal is to evaluate the interaction of anthropogenic pollutant plumes and a Mediterranean oak ecosystem (with well known isoprene emissions) in a dynamical idealized situation. Two different chemical anthropogenic sources have been tested for different distances between TOWN and FOREST area (from d_1 – the shortest to d_4 – the longest).

For the Marseille area, the anthropogenic emission is characterized by a relatively high VOC/NO_x ratio (around 5 ppbC/ppb at the emission). The impact of forest emissions on ozone formation in the pollutant plume is more important in proximity of the

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TOWN area. In such a configuration, the contribution of biogenic ozone can reach 28% of the ozone maximum concentration, and 10% of the total amount of ozone present in the plume at the end of the afternoon. Due to the specificity of the emissions, the anthropogenic plume of the Marseille area rapidly evolves towards a NO_x sensitive chemical regime. The biogenic impact strongly decreases when the TOWN-FOREST distance increases to become insignificant for distances >70 km and a wind speed of 4 m s^{-1} .

The biogenic impact is sensible for larger TOWN-FOREST distances for the Martigues source which has a VOC/NO_x ratio of around 2 ppbC/ppb at the emission. The ozone production from biogenic precursors is limited by high NO_x concentrations in the morning and with reduced TOWN-FOREST distances. The larger biogenic impact on maximum ozone concentrations is found for the d_3 distance (21%) on the morning, i.e. for a TOWN-FOREST distance around 70 km and a wind speed of 4 m s^{-1} . As the NO_x concentration decreases during the day, the biogenic maximum impact zone shifts towards the TOWN area. The biogenic impact is then larger because isoprene emissions are more important around midday (27% of the maximum ozone concentration due to biogenic precursors for the distance d_1). Considering the total amount of ozone in the plume, the maximum production due to biogenic precursors is observed for the smallest TOWN-FOREST distance (d_1) with a contribution of 15% due to biogenic precursors at the end of the day. Contributions calculated for longer distances are not negligible (see Sect. 4.5.2).

In this idealized study, the horizontal wind velocity was fixed to 3 m s^{-1} . The different results obtained for the d_1 to d_4 distances must account for this velocity. A change of the wind velocity would affect the advection time of primary compounds on forest area and thus the time scale of interaction between polluted plume and forest. Concentration fields would be equally affected due to different conditions of diffusion and residence time of air masses above the forests. An idealized simulation using simplified landscape and atmospheric dynamics is an interesting tool to better isolate processes occurring at regional scales, and to carry out sensitivity test for the evaluation of the

impact of biogenic sources. This kind of simulation can be useful if we focus on well-
documented areas, eventually with regulatory policies. Furthermore, developments
presented here are designed for Meso-NH-C real case regional simulations. In such
configuration, the BVOCEM emission scheme will be used in connection with regional
emission potential inventories in future works.

Acknowledgements. Many thanks to J. Fuentes and C. Geron for providing experimental data
of isoprene and surface fluxes above the boreal and Duke forests. We would also like to thank
S. Rambal, P. Bouchou and A. Fotiadi for there the work done in the Mediterranean forest.

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Table 1. Main characteristic of isoprene emitting ecosystems considered for validations. Details on isoprene emissions potential estimations: (a) Obtained considering a species emission factor for *populus* equal to $50 \mu\text{g g}_{dw}^{-1} \text{h}^{-1}$ (Kesselmeier et al., 1999). The resulting value close to the direct estimation of Fuentès et al. (1999) who obtained $28 \text{ nmol m}^{-2} \text{ s}^{-1}$ ($7000 \mu\text{g m}^{-2} \text{ h}^{-1}$) from seasonal measurements on the BOR site; (b) Obtained from in situ vegetation screenings giving a mean species emission factors equal to $36,7 \mu\text{g g}_{dw}^{-1} \text{h}^{-1}$, (Guenther et al., 1994); (c) Obtained from in-situ vegetation screening over 82% (in term of *LAI*) of present species (Serça et al., 2001); (d) Obtained from an emission factor equal to $22,75 \mu\text{g g}_{dw}^{-1} \text{h}^{-1}$ for *Quercus pubescens* (Hewitt et al. (1996), see also text). *Juniperus* understorey is considered as non emitter.

Ecosystem	BOR	DUK	TROP	MED
Situation	53,7°N; 106,2°W	35°58'25"N; 79°06'55"W	2° N; 16°E	43°39'N; 3°41'E
Characteristics	Boreal forest, Homogeneous canopy with understorey cover	Temperate forest, Dense canopy, flat terrain Duke University instrumented site.	Dense tropical canopy EXPRESSO instrumented site (Delmas et al., 1999)	Mediterranean forest Flat terrain (8000 ha) Light canopy
Dominant species	<i>Populus tremuloides</i> <i>Populus balsamifera</i> (<8%) non emitting understorey	<i>Quercus sp</i> Strong emitters	Large variability	<i>Quercus pubescens</i> (85%) <i>Juniperus</i> (understorey)
Height (m)	22	28-32	45	6
<i>LAI</i> ($\text{m}^2 \text{ m}^{-2}$)	5.6 (total), 2.4 (emitters)	5.2	6.3	2.4 (total)
Dry leaf biomass ($\text{g}_{dw} \text{ m}^{-2}$)	144 (emitters)	472	-	228
Isoprene <i>EP</i> ($\mu\text{g m}^{-2} \text{ h}^{-1}$)	7000 (a)	17300 (b)	4443 ©	5185 (d)
Micro-meteorological forcing	Above canopy radiations, wind, air temp. and hum. Net rad., sens and lat. Heat fluxes	Above canopy radiations, wind, air temperature and humidity. Net rad., sens. And lat. Heat fluxes	Above canopy radiations, wind, air temperature and humidity. Sens. And lat. Heat fluxes	Above canopy radiations, wind, air temperature and humidity. Sens. And lat. Heat fluxes
Isoprene fluxes method	Flux-gradient method	Relaxed Eddy Accumulation	Relaxed Eddy Accumulation (Rq. Gaps in measurements)	Fast isoprene sensor + covariance method
Period of measurement	Days 203, 207 and 215 August 1994	8 successive days June, 1996	17-23 March 1996	28 June – 2 July 2000
References	Fuentès et al., 1999	Guenther et al., 1994. Geron et al., 1997. Guenther et al., 1998.	Klinguer et al., 1998 Serça et al., 2001	-

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Table 2. Correlation coefficient for the four sites considered for validations.

Ecosystem	BOR	DUK	TROP	MED
Correlation coefficients (%)	0,89	0,91	Not enough experimental data	0,88

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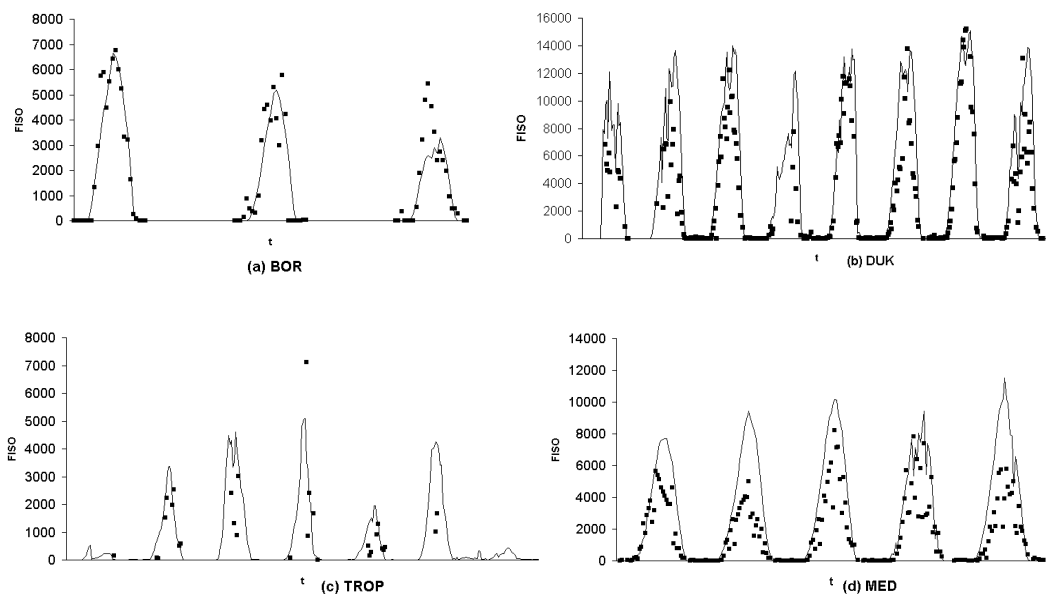


Fig. 1. Simulated isoprene emission fluxes (solid lines) in $\mu\text{g m}^{-2} \text{h}^{-1}$ versus measured emission fluxes (dots) for four contrasted ecosystems. **(a)** BOREal forest for three separate days; **(b)** DUKe forest temperate ecosystem for 8 consecutive days; **(c)** TROPical forest for 6 consecutive days; **(d)** MEDiterranean forest for five consecutive days. See text and Table 1 for descriptions of these ecosystems.

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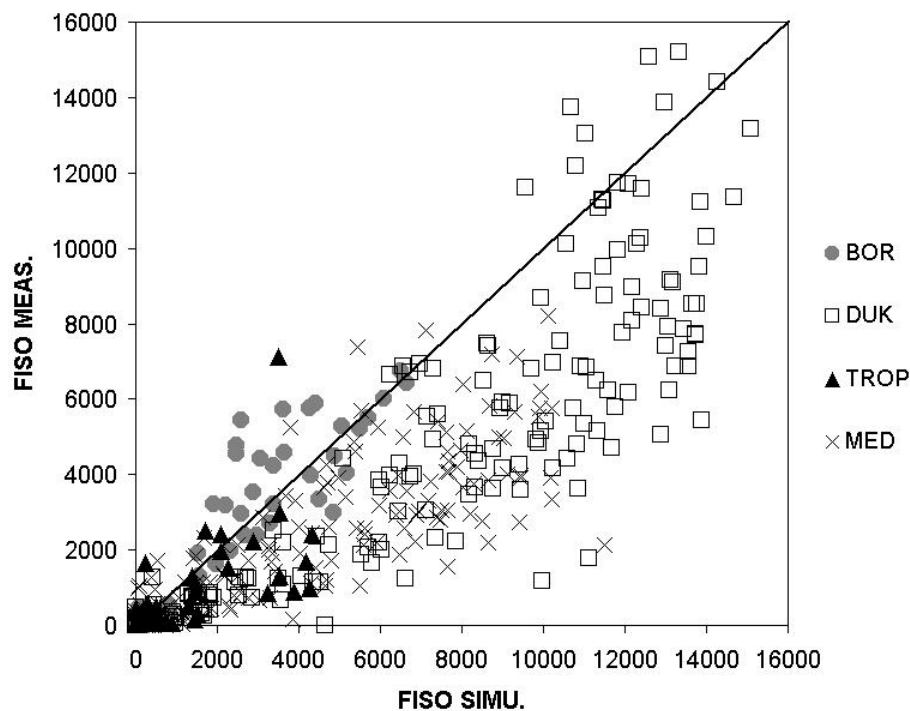


Fig. 2. Global scatter plot of model results versus isoprene flux measurement in the four ecosystems (see Fig. 1 legend).

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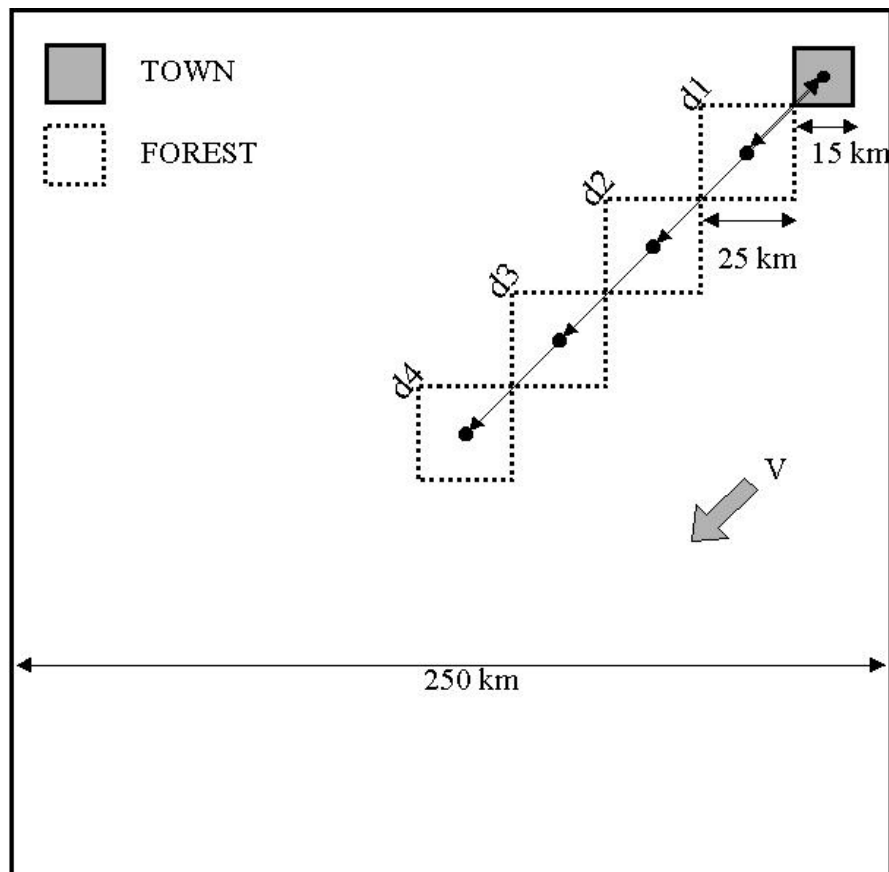


Fig. 3. Schematic configuration of the idealized domain of simulation to study the interactions between of the anthropogenic plume coming from TOWN area and isoprene emission from the FOREST area. d_1 to d_4 represent different downwind distances between TOWN and FOREST.

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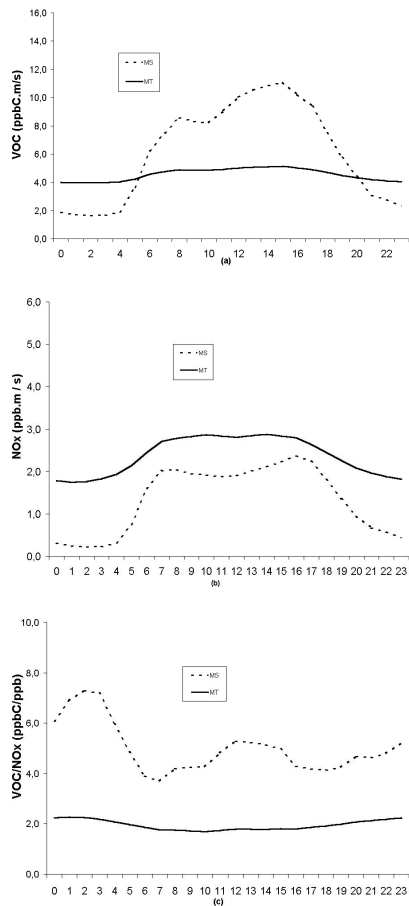


Fig. 4. Chemical characteristics of the anthropogenic sources for the TOWN area. **(a)** Diurnal evolution of VOC fluxes for Marseille (*MS*) and Martigues (*MT*) scenarios; **(b)** Diurnal evolution of NO_x for *MS* and *MT* scenarios; **(c)** Corresponding VOC/NO_x ratios.

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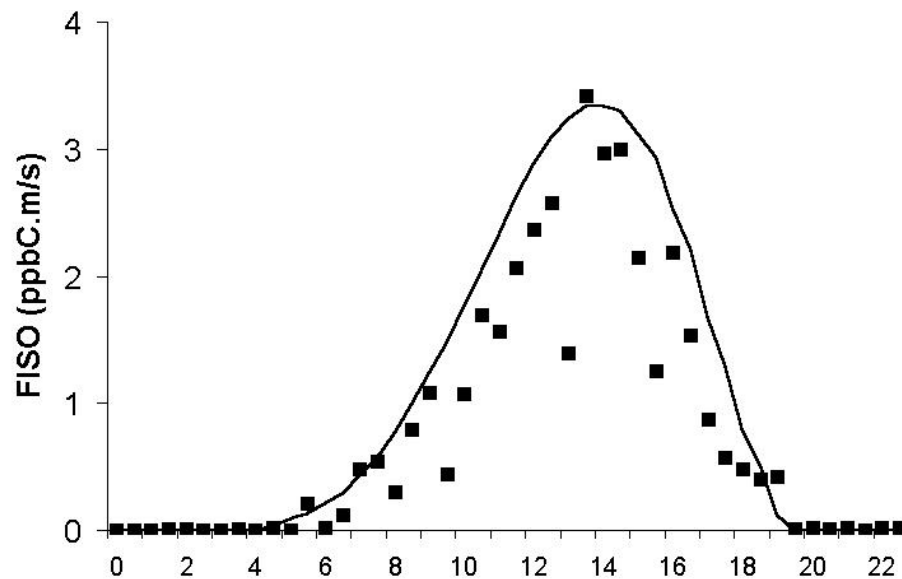


Fig. 5. Isoprene emission flux above the FOREST area for the 3-D simulation. Dots represent experimental data collected during the Mediterranean field campaign (see text, Sect. 3).

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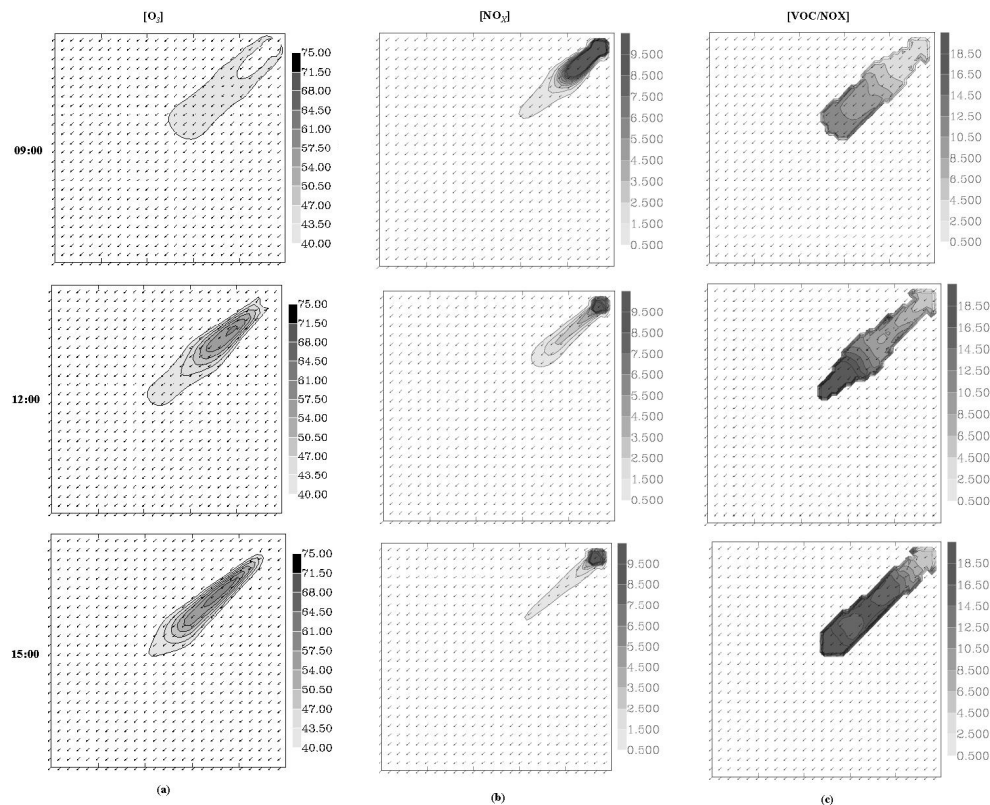


Fig. 6. Reference simulations without biogenic emissions for the Marseille case (*MS-NOBIO*). Horizontal section (at 10 m above ground) are given at 09:00 UT, 12:00 UT and 15:00 UT. **(a)** and **(b)**: Ozone and NO_x concentration fields (in ppb), **(c)** VOC/ NO_x ratio.

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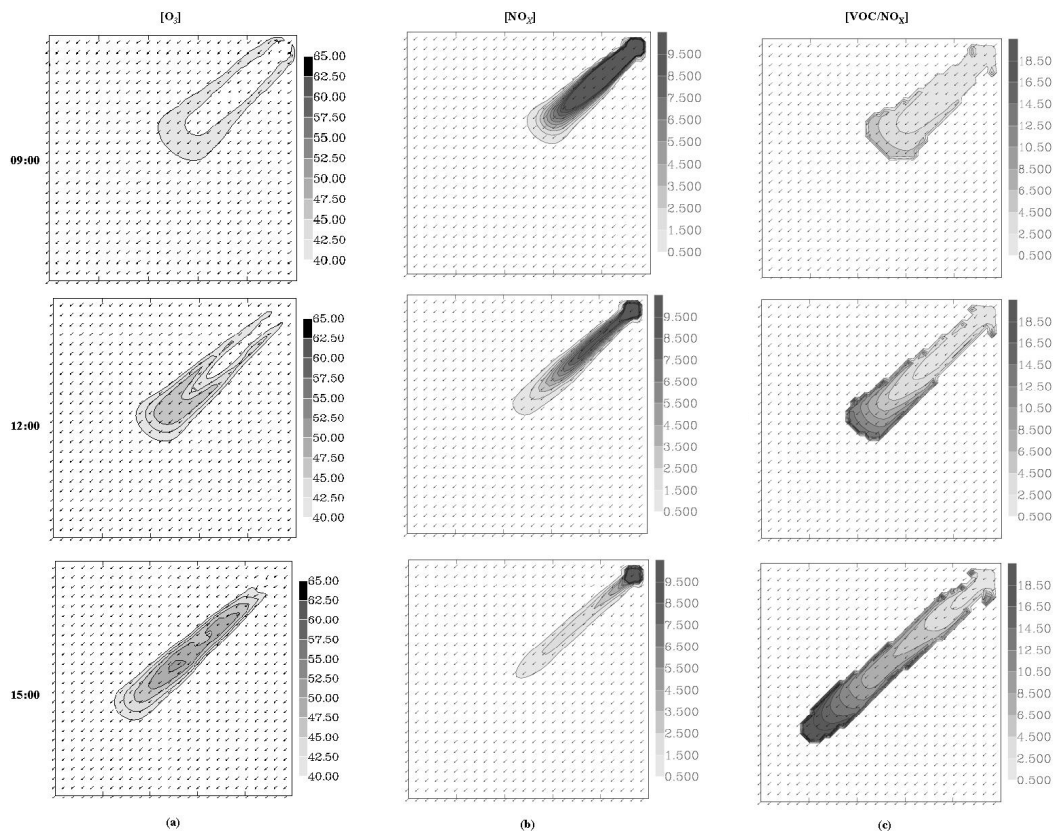


Fig. 7. As Fig. 6, for Martigues case (*MT-NOBIO* simulation).

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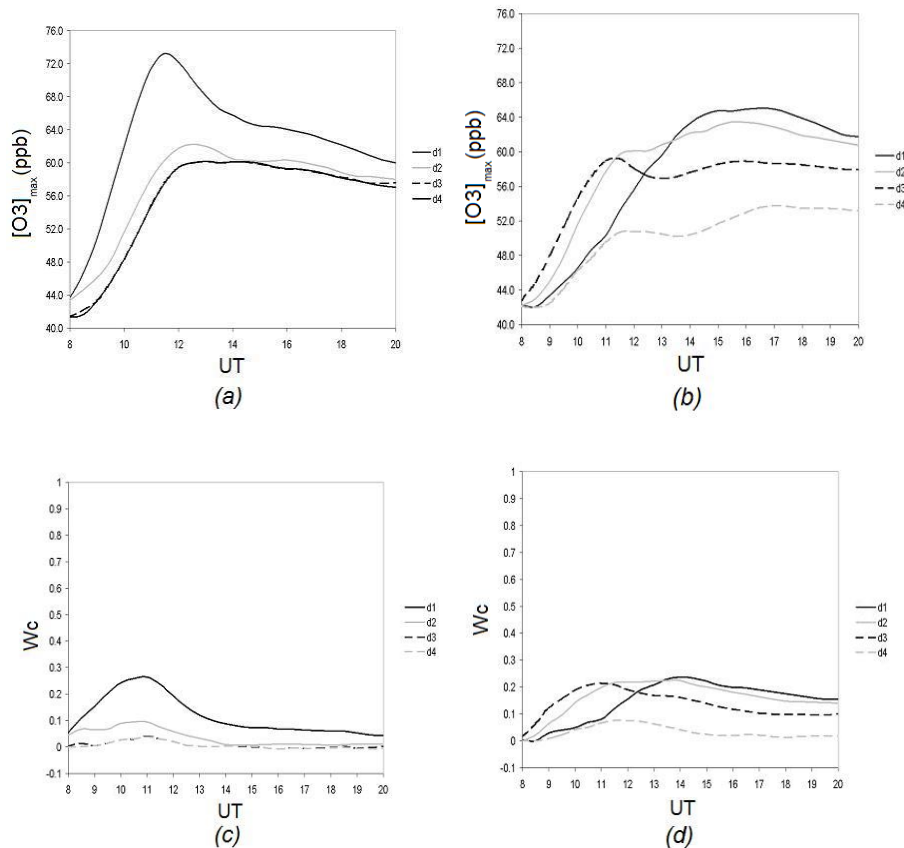


Fig. 8. Results of simulation taking into account biogenic emission over the FOREST area. **(a)** Evolution of the ozone maximum concentration in the *MS* case for different downwind distances between TOWN and FOREST (d_1 to d_4); **(b)** Same as (a) for the *MT* scenario; **(c)** Evolution of the relative contribution of isoprene emission to ozone maximal concentration W_c (see definition in the text) in the *MS* case; **(d)** Same as (c) for the *MT* scenario.

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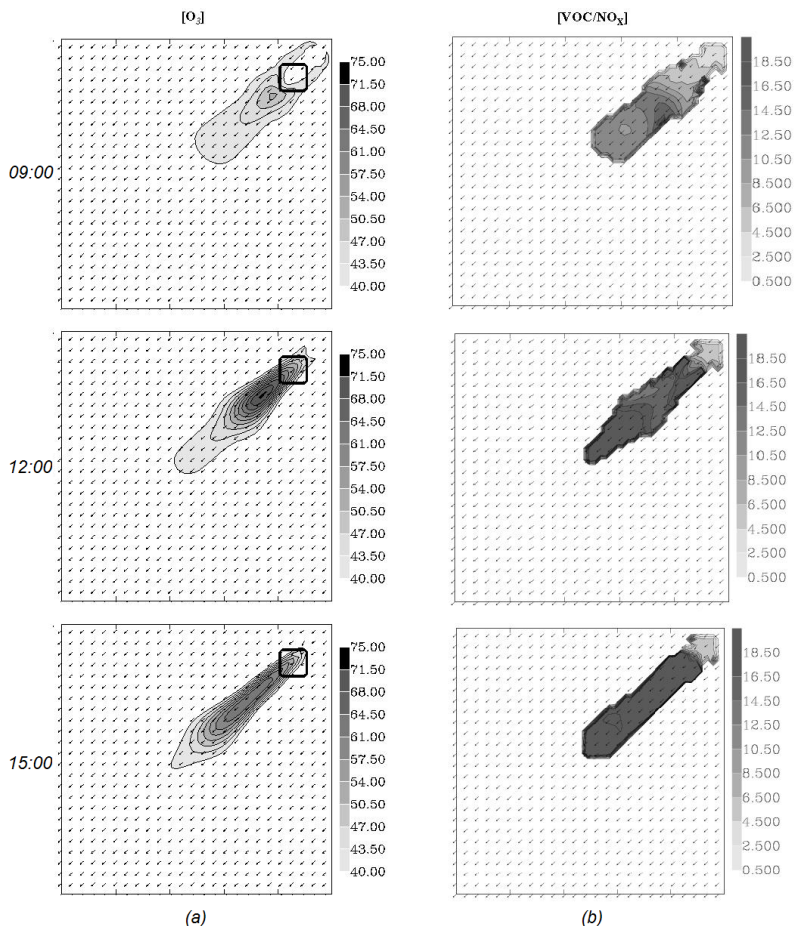


Fig. 9. Simulation results taking into account biogenic emission for the *MS* case with the d_1 TOWN-FOREST distance. Horizontal section (at 10 m above ground) are given at 09:00 UT, 12:00 UT and 15:00 UT. **(a)** Ozone concentration field (ppb); **(b)** VOC/NO_x ratio (ppbC/ppb).

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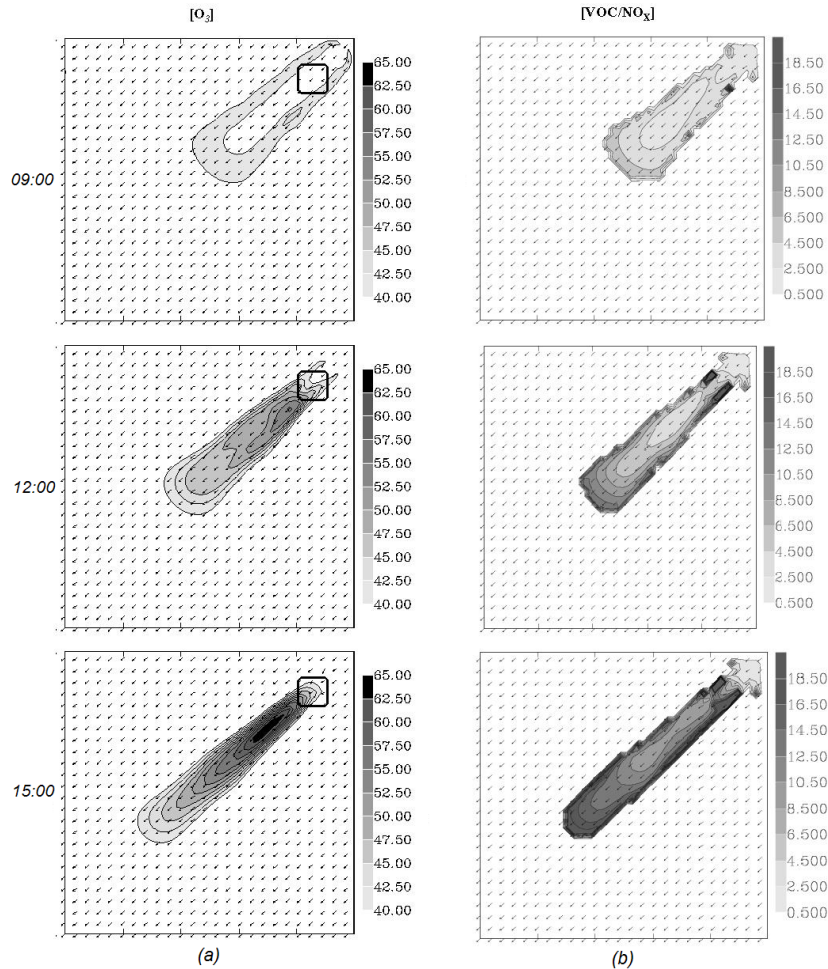


Fig. 10. Same as Fig. 9 for the MT scenario.

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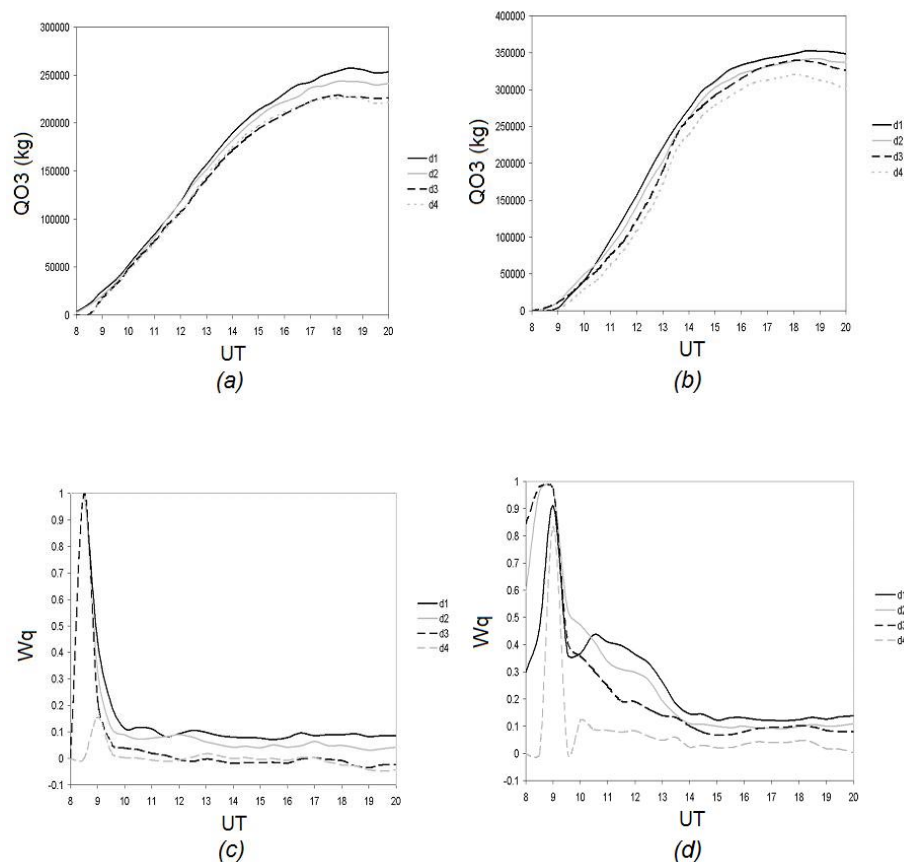


Fig. 11. (a) Simulation results taking into account biogenic emission over the FOREST area. (a) Total ozone amount in the plume for the *MS* situation, function of d_1 to d_4 TOWN-FOREST distances; (b) Same as (a) for *MT* scenario; (c) Evolution of the relative contribution of isoprene emission to the total ozone amount W_Q (see definition in the text) in the *MS* case; (d) Same as (c) for the *MT* scenario.

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