BACCHUS

Impact of Biogenic versus Anthropogenic emissions on Clouds and Climate: towards a Holistic UnderStanding

Collaborative Project

SEVENTH FRAMEWORK PROGRAMME
ENV.2013.6.1-2

Atmospheric processes, eco-systems and climate change
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Responsible	Jurgita Ovadnevaite
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Internal reviewer:	Maria Kanakidou

Executive summary

This deliverable is related to Question 1a of the project: ‘What are the levels and properties of the natural background aerosol in different key regions and how can they be used to constrain estimates of pre-industrial conditions so as to quantify the anthropogenic aerosol effect since pre-industrial times?’

In particular, the aim was to improve the **marine aerosol**, including organics, and the **terrestrial biogenic aerosol**, and their precursor, source parameterizations for use in CTMs and ESMs. D2.1 focuses on the sea-spray source function and on primary biogenic aerosol from terrestrial sources. The developed parameterizations are related to indicators of “biological activity” (chlorophyll, terrestrial vegetation dynamic) and are suitable for different environments. The effects of climate change and changes in atmospheric CO_2 levels on fire conditions, amount of combustible litter and vegetation **wildfire emissions** were assessed and provided for ESM studies in WP4. The existing parameterizations of these natural sources were improved based on extensive interpretation of observations in WP1. For primary biological aerosol particles (PBAPs) extensive measurements of fluorescent biological particles from Task 1.3 has been used.

Summary of results

1. Marine aerosols source parameterization.

Parameterizations for the marine sources of aerosols are presented as sea spray source function (SSSF) with marine primary organic aerosol (POA) source function parameterization.
The SSSF does not include organic matter which is a very important part of the sea spray aerosol, in particular for the smallest particles (Facchini et al., 2008), which can act as cloud condensation nuclei. Therefore, we suggest using the OSSA-SSSF (see ***Sea spray source function***) to obtain the total sea spray aerosol fluxes and combine it with a parameterization developed by Rinaldi et al. (2013) to derive the organic fraction (see Marine POA source function parameterization).

1.1. Sea spray source function (NUIG).

Submicron sea spray aerosol fluxes derived from particle number concentration measurements at the Mace Head coastal station, on the west coast of Ireland, were used together with open-ocean eddy correlation flux measurements from the Eastern Atlantic Sea Spray, Gas Flux, and Whitecap (SEASAW) project cruise, to derive a sea spray source function (OSSA-SSSF). The OSSA-SSSF covers the dry diameter range from 15 nm to 6 µm. The source function has been parameterized in terms of five lognormal modes and the Reynolds number (Eq. 1) instead of the more commonly used wind speed, thereby encapsulating important influences of wave height, wind history, friction velocity, and viscosity. This formulation accounts for the different flux relationships associated with rising and waning wind speeds since these are included in the Reynolds number (Re_{H_w}) (Eq. 2). Furthermore, the Reynolds number incorporates the kinematic viscosity of water, thus the SSSF inherently includes dependences on sea surface temperature and salinity.

$$\frac{\mathrm{dF}}{dlogD} = \sum_{i=1}^{\mathrm{n}} \frac{F_i(Re_{Hw})}{\sqrt{2\pi} ln\sigma_i} \exp\left(-\frac{1}{2} \left(\frac{\ln\left(\frac{D}{CMD_i}\right)}{ln\sigma_i} \right)^2 \right) \qquad (1)$$

Here dF/dlog$D$ is the size dependent SSA production flux, i- mode number, $F_i(Re_{Hw})$ –the flux for mode i, D –dry particle diameter, σ-geometrical standard deviation, and CMD – count median diameter.

$$Re_{H_w} = \frac{C_d^{\frac{1}{2}} U_{10} H_s}{\nu_w} \qquad (2)$$

Here C_d is the drag coefficient, U_{10} is the 10 m wind speed, H_s is the significant wave height and ν_w is the viscosity of water.

The temperature dependence of the resulting SSSF is similar to that of other in-situ derived source functions (Jaeglé et al., 2011) and results in lower production fluxes for cold waters and enhanced fluxes from warm waters as compared with SSSF formulations that do not include temperature effects, such as Gong (2003) (Figure 1).

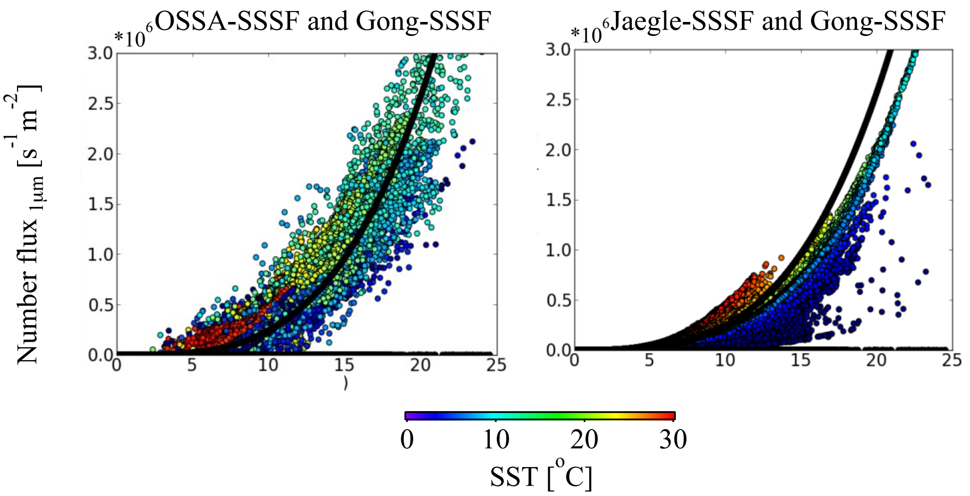


Figure 1. Global instantaneous (2 Jan 2006, 00 UTC) SSA number flux (for particles 0.07<D< 1µm); Left panel: computed using the OSSA-SSSF (colour circles) overlapped with black dots representing fluxes computed using the Gong (2003) SSSF; Right panel: computed using the Jaeglé et al. (2011) SSSF (colour circles) overlapped with black dots representing fluxes computed using the Gong (2003) SSSF; Colours represent sea surface temperature (SST).

This source function shows and accounts for the distinct forcing effects on the different particle size ranges, resulting in different flux distributions for a particular Re_{Hw} (Table 1).

Table 1. Lognormal parameters for the SSSF parameterization. For each mode, a geometric standard deviation (σ_i), count-median diameter (CMD_i), and the total number flux (F_i) as a function of Reynolds number (Re_{Hw}) are given.

i	σ_i	CMD_i	$F_i(Re_{Hw})$
1	1.37	0.018	$104.5(Re_{Hw}-1*10^5)^{0.556}$
2	1.5	0.041	$0.0442(Re_{Hw}-1*10^5)^{1.08}$
3	1.42	0.09	$149.6(Re_{Hw}-1*10^5)^{0.545}$
4	1.53	0.23	$2.96(Re_{Hw}-1*10^5)^{0.79}$
5	1.85	0.83	$0.51(Re_{Hw}-2*10^5)^{0.87}$

For more details on the source function see (Ovadnevaite et al., 2014).

1.2. Marine POA source function parameterization (NUIG, CNR-ISAC)

The relation between the organic fraction of sea-spray aerosol (OM_{SS}) and oceanic biological activity over the Western North Atlantic Ocean has been investigated by Rinaldi et al. (2013), through offline aerosol chemical composition measurements, performed at Mace Head, and state-of-the-art ocean colour products from satellite observations. As a result of this activity, an improved marine primary organic aerosol (POA) source function for large scale models was developed. This source function allows for the partitioning of the sea-spray mass generated by traditional production schemes, as a function of wind speed, into organic and inorganic components, using chlorophyll-a concentration (Chl-a) and wind speed (U_{10}) as input parameters, according to the following equation:

$$OM_{SS} = (56.9 \text{ x Chl-a}) + (-4.64 \text{ x } U_{10}) + 40.9$$

The improved correlation obtained by Rinaldi et al. (2013), with respect to previous attempts (e.g., Vignati et al., 2011), is due to the use of more refined satellite data (deriving from the merging of SeaWiFS, MODIS and MERIS raw data) and to an improved satellite data treatment (MSSA algorithm).

The novel source function by Rinaldi et al. (2013) has successfully been applied by Partanen et al. (2014), resulting in a global estimated POA flux of 1.1 Tg yr^{-1} (sensitivity range 0.5–1.8 Tg yr^{-1}). This value is well in the range of 0.1–11.9 Tg yr^{-1} simulated by Gantt et al. (2012), who compared six different ways to estimate the organic mass fraction of sea spray aerosol emissions.

Within **BACCHUS**, the parameterization of Rinaldi et al. (2013) was updated by reanalysing observations in WP1. A multi-year dataset spanning the period from 1st January 2009 till 30th September 2011 and covering three phytoplankton bloom periods of OM, sea salt and OM_{SS} (derived from Aerosol Mass Spectrometry measurements) in conjunction with a daily-timescale reanalysis dataset of satellite-derived biological proxy outputs (i.e. *Chl-a* and Net Primary Productivity - NPP) has been used to corroborate the relation between the organic mass fraction of sea-spray and the oceanic biological activity. The high time and mass resolution AMS dataset provided a better control of anthropogenic contaminations and allowed an investigation of a time dependant correlation. The shortest timescale analysed using the satellite data was one day and led to a correlation coefficient between Chl-a and OM_{SS} of R=0.67 (Figure 2). Moreover, Figure 2 illustrates that as the timescale was lengthened to one week, the correlation increased to 0.76, and up to 0.85 for a month-long timescale while NPP exhibited a correlation of 0.93, again for a month–long timescale, confirming that OMss is neither constant nor independent of biological activity.

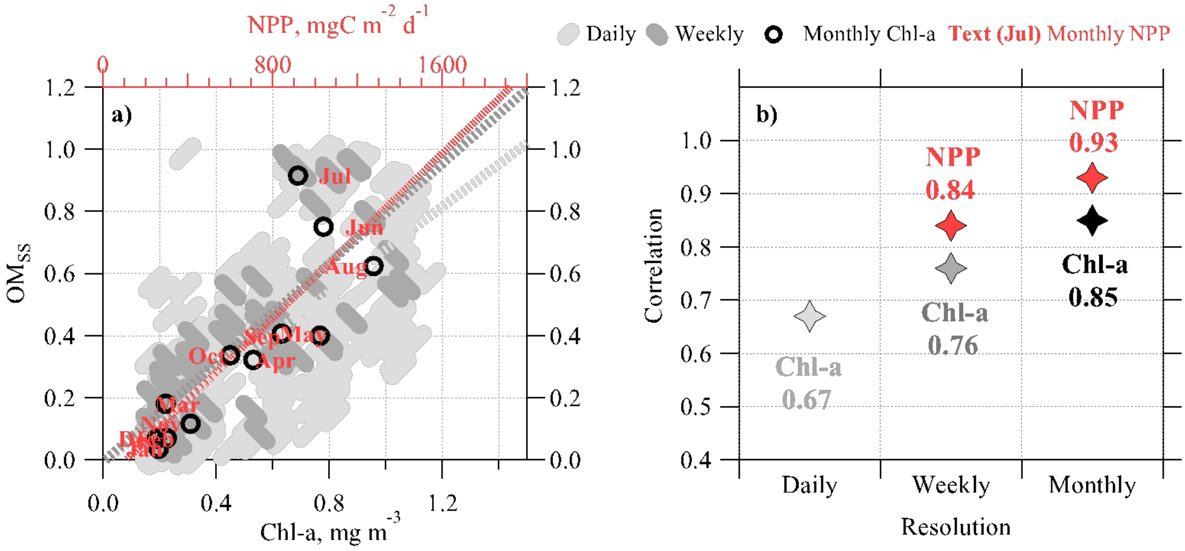


Figure 2. (a) Scatter plot of sea spray OM_{SS} as a function of Chl-a for daily, weekly and monthly temporal resolutions and NPP for monthly temporal resolution; (b) correlation coefficient as a function of temporal resolution of biological indicators.

The aforementioned correlation maximum for monthly average concentrations, combined with the observations of a 26-day "boom-bust" biological cycle timescale associated with bloom dynamics as recently illustrated by Lehahn et al. (2014), suggests that the maximum correlation occurs at a timescale longer than 8 days and perhaps the peak in OM_{SS} is more associated with the demise of the bloom rather than any growth phase. In particular, these results are consistent with the time lag between the initiation of the phytoplankton bloom (generally assumed to occur when Chl-a concentrations pass the threshold of 1 µg L^{-1}), the increase of prokaryotes that can exploit the phytoplankton exudates and the ability of viruses to infect living cells (Wommack et al., 2000).

This suggestion is supported here through examination of the correlation coefficient frequency function for NPP, for time lags of 0, 8, 16, and 24 days. It was observed that the correlation between NPP and OMss increases as the time lag is increased from 0 to 16-24 days. This suggests a maximum correlation between OMss and NPP for the latter third of a typical bloom life span, or, in other words, maximum correlation is seen for bloom demise rather than bloom growth (For more details see O'Dowd et al. (2015)).

The new dataset confirms the observations of Rinaldi et al. (2013) on the OM_{SS} vs. Chl-a relationship and delay time, i.e. the time lag between the observation of the Chl-a bloom and the enhancement of the OM_{SS}, although, this time lag seems to be inconstant and dependent on the specific bloom conditions.

In conclusion, the source function by Rinaldi et al. (2013) is proposed to the BACCHUS community as the reference marine POA parameterization for large scale models. However, future users of the function are invited to consider carefully the caveats for its use, discussed in the Rinaldi et al. (2013) and the paper by O'Dowd et al. (2015).

2. Parameterizations for the terrestrial sources of aerosols and their precursors

2.1. Emission parameterizations of primary biological aerosol particles

Primary biological aerosol particles (PBAP) are aerosols that carry living organisms or are released by them (Ariya and Amyot, 2004). Such aerosols can be viruses, bacteria, fungal spores, plant debris, pollen and their sizes span from the sub-to the super micron levels. PBAPs account for a significant amount of aerosol mass with global emission estimates spanning over one order of magnitude. They are present both over land and over the oceans. Primary biological aerosol particles can influence the cloud formation by acting as cloud condensation nuclei (CCN) and/or ice nuclei (IN).

2.1.1. Development of a Global PBAP Emission Parameterization based on in-situ FBAP Measurements (MPI-C).

Here, we are developing a parameterization method of global PBAP emissions in the framework of BACCHUS, based on in-situ FBAP (fluorescent biological aerosol particles) measurements over multiple sites across the world. Some major improvements of this work are summarized as below:

First, the in-situ FBAP measurements from Ultraviolet Aerodynamic Particle Sizer (UV-APS) and Waveband Integrated Bioaerosol Sensor (WIBS) instruments over the multiple sites are used to constrain the global parameterization scheme. These measurements provide high resolution and consistent data of the atmospheric abundance of PBAP over various terrestrial categories in different continents and seasons, and will extend the limited in-situ measurements in previous studies.

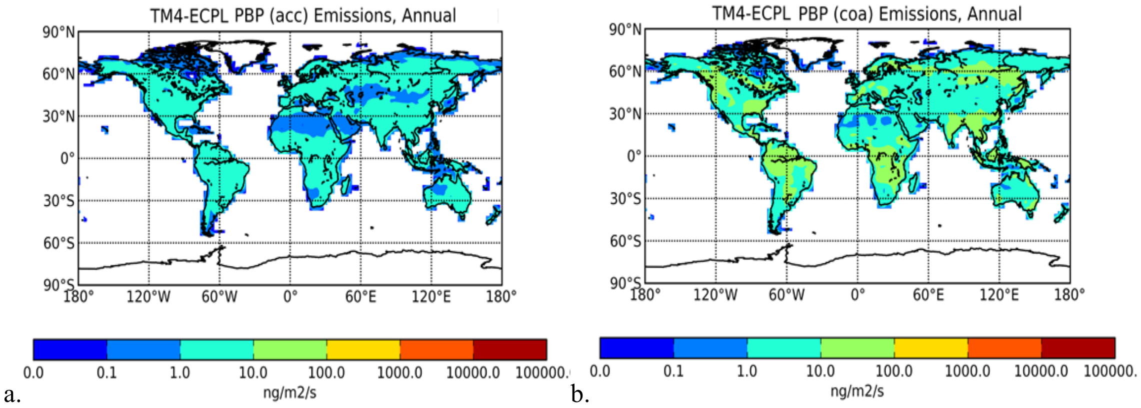
Second, latest progress in field measurements shows that more parameters e.g., precipitation, are needed to account for the observed emission PBAP (Huffman et al., 2013; Schumacher et al., 2013). The increasing PBAP measurements during rainfall observed by field measurements is thought to be related to mechanical ejection from terrestrial surfaces as a result of rain droplet splash. Thus, an impulse term is introduced in the emission parameterization of PBAP to take into account the effect of precipitation.

Third, the water-surface PBAP emissions will be quantified to extend the emission estimates of PBAP to the entire earth surface. The chlorophyll concentration derived from the SeaWiFS satellite instrument will be used to provide the biological activities at the surface layer of the oceans.

The estimated global PBAP emissions incorporated into a global chemical transport model and a coupled climate model will be used to simulate the PBAP concentrations that will be then compared to the in-situ FBAP measurements for further improvements. We are quantitatively examining the contribution of PBAP to the atmospheric aerosol compositions over different regions, and investigate their roles in the in-cloud micro-physical and chemical processes as a source of CCN and IN.

2.1.2. Improvement of PBAPs emission parameterization in a global CTM (UOC)

PBAPs emission parameterizations discussed in literature (see references below) for various scale models are being tested in the TM4-ECPL global chemistry transport model. Both fine and coarse mode particle emissions are taken into account. In addition to the leaf area index that is driving this emissions variability (Figure 3), the variability with meteorological conditions and ecosystem type is important.



a. b.

Figure 3. Annual mean emission distribution of PBAP in TM4-ECPL model parameterized based on leaf area index a) fine mode b) coarse mode (simulated as in Kanakidou et al., 2012).

Burrows et al. (2009) provided near surface bacteria fluxes for 4 different ecosystem types used by Sesartic et al. (2012). Sesartic and Dallafior (2011) compiled similar information for fungal spores for 6 different ecosystems. Heald and Spracklen (2009) developed a parameterization of fungal spore fluxes, used by Hoose et al. (2010) based on leaf area index and specific humidity and further improved by Hummel et al (2015). These parameterisations are being tested in the model and will be evaluated against observations compiled in the project.

2.2. The parameterization of biogenic volatile organic carbon (BVOC) emissions in LPJ-GUESS (KIT).

Current studies on the atmospheric impact of BVOC´s are overwhelmingly based on one empirically-derived model (Guenther et al., 1995; Guenther et al., 2012). The BVOC module in the global dynamic vegetation model LPJ-GUESS is an alternative (semi-)process-based approach, linking emissions to the photosyntethic activity of the vegetation. The BVOC emission module combines the process-based leaf level emission model from Niinemets et al. (2002;1999) with the

LPJ-GUESS vegetation model as described in Arneth et al. (2007) for isoprene and in Schurgers et al. (2009) for monoterpenes. In this model, the isoprene and monoterpene production in the leaf is linked to the electron transfer rate of carbon assimilation, which supplies important metabolites for BVOC synthesis in the chloroplast. Monoterpenes can be directly emitted just as isoprene or (partially) stored for some time. In the model it is assumed that half of the monoterpenes are stored and released as a function of leaf temperature.

Adopting the concept of plant functional types (PFT) specific emission capacities (Guenther et al., 1995), the emission rates for different PFTs are scaled such that under standard conditions (30°C, photosynthetic active radiation (PAR) of 1000 µmol $m^{-2}s^{-1}$ and ambient CO_2) the emissions per unit dry leaf biomass are equal to those given by Arneth et al. (2007a). The model takes into consideration the direct and indirect process-response of BVOC emissions to changing climate and CO_2 concentration, including responses at the leaf as well as at the ecosystem scale. These include the CO_2 inhibition on leaf production of isoprene and monoterpene.

Until now, all monoterpenes were considered by using a single emission factor without distinction between different chemical species of monoterpenes – despite of their very different atmospheric impacts. Here we adapted the LPJ-GUESS BVOC module to include the main monoterpene species. Furthermore, we adapted the monoterpene storage fraction (the fraction of monoterpenes, which is not directly emitted, but stored internally and emitted slowly depending on a temperature function) which is currently a fixed 50% to a monoterpene-species dependent storage fraction. To do so, the total monoterpene emission factor currently implemented in LPJ-GUESS was maintained. The partitioning of the resulting emissions into different chemical species is performed based on a recent literature review by P. Messina (LSCE, France; in prep.), summarizing the contributions of the chief eight monoterpene species (and one "rest monoterpene" group) to the total emissions for each PFT. The monoterpene storage factors were taken from Guenther et al. (2012), they differ for each monoterpene species but are the same for each PFT. Where no storage data was available 50% storage was assumed. The resulting spatial distribution in emissions for two contrasting species (with contrasting SOA formation) is given in Figure 4.

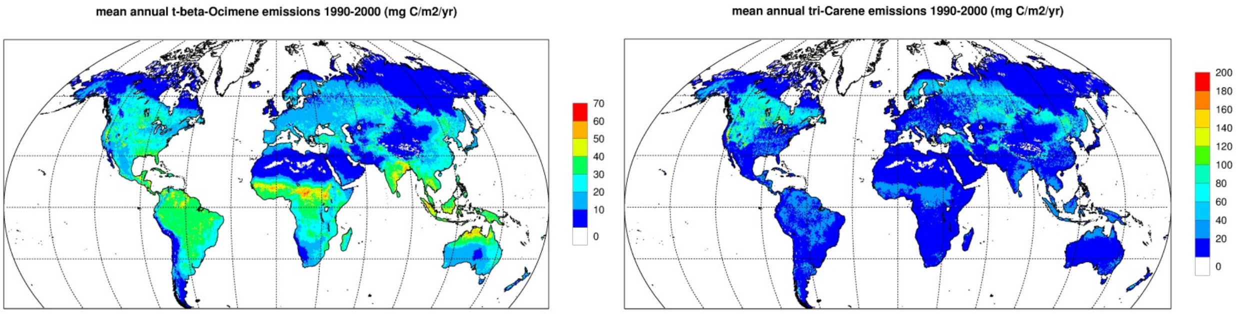


Figure 4: Mean annual emissions (1990-2000) for the 2 monoterpene species with different spatial distributions: ***left panel*** t-beta-Ocimene; ***right panel*** tri-Carene.

2.3. Fire emissions (KIT)

Multiple fire models have been developed during the last decades. However, their performance has never been evaluated structurally. The current fire module in LPJ-GUESS was found to overestimate both burned area and fire emissions strongly, unsuitable to model fire emissions globally. Therefore the fire model SIMFIRE (Knorr et al. 2014) as integrated in the LPJ-GUESS global vegetation model. SIMFIRE is a simple but non-linear model of fractional burned area and was developed by Knorr et al. (2014), optimized against observed burned-area products. LPJ-GUESS-SIMFIRE produces offline monthly emission fields and can be used to estimate fire emissions both for past climate and future scenarios. Details on the implementation of SIMFIRE in LPJ-GUESS can be found in Deliverable 4.1.

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Changes with respect to the DoW

none

Dissemination and uptake

The improved marine source parameterizations has been already available for modelling efforts outside this project and they will be implemented in the ESMs within the BACCHUS framework (Task 4.1); a paper on the POM parameterization improvements has been submitted to *Scientific Reports*.

PBAP Emission Parameterizations will be implemented in global models (Task 4.1).

The monoterpene speciation has been developed in concordance with the requirements in Task 4.1 as off-line inputs/ alternative inputs for the different ESM´s (see deliverable 4.1).

LPJ-GUESS-SIMFIRE implementation is related to deliverable 4.1.