

Project Number 282910

ÉCLAIRE

**Effects of Climate Change on Air Pollution Impacts and Response
 Strategies for European Ecosystems**

Seventh Framework Programme

Theme: Environment

**D7.3 Report on effects of in-canopy BVOC and NO emissions on in-canopy
 O3 and POD estimates.**

Due date of deliverable: **01/06/2015**

Actual submission date: **30/09/2015**

Start Date of Project: **01/10/2011**

Duration: **48 months**

Organisation name of lead contractor for this deliverable :

Norwegian Meteorological Institute

Project co-funded by the European Commission within the Seventh Framework Programme		
Dissemination Level		
PU	Public	X
PP	Restricted to other programme participants (including the Commission Services)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	

Executive Summary

The one-dimensional ECLAIRE Ecosystem Surface eXchange (ESX) model developed jointly with WP4 has been coupled in offline mode with the EMEP MSC-W chemical transport model, and used to investigate the effects of in-canopy emissions of biogenic volatile organic compounds (BVOC) and of soil-NO emissions on in-canopy O₃ and phyto-toxic ozone dose (POD) estimates at a number of sites across Europe.

Although this model system needs further evaluation once the ECLAIRE field-data analysis has been finished, the ESX results suggest that calculated POD values are remarkably stable given varying biogenic emissions. This is largely due to the fact that POD values are calculated from top-of-canopy O₃ values, whereas the main effects of the in-canopy chemistry are to modify in-canopy values, especially in the lowest layers. The biogenic emissions do have significant effect on total O₃ deposition though in these tests though, with differences of around 7% between high and low emission scenarios.

1 Objectives

The objective of D7.3 is to report on the effects of in-canopy emissions of biogenic volatile organic compounds (BVOC) and of soil-NO emissions on in-canopy O₃ and phyto-toxic ozone dose (POD) estimates across Europe.

2 Activities

In order to address issues concerning in-canopy chemistry across Europe, we have developed a new 1-dimensional model, the ECLAIRE Ecosystem Surface eXchange (ESX) model, and enabled a link through offline nesting to the European scale EMEP MSC-W chemical transport model (Simpson et al. 2012). The basic structure of the ESX model has been introduced in Simpson and Tuovinen (2014) and Deliverable D4.4, and a documentation paper is in preparation. The ESX model is unique in that it was designed from the start to satisfy the needs of both the analysis of field-scale measurements and large-scale chemical transport modelling. The model is also closely tied to the EMEP MSC-W model system, and for this report we drive ESX for different locations across Europe with the EMEP model.

Although ESX can be run in stand-alone mode or Lagrangian modes, we present here results from a one-way nesting setup, whereby boundary conditions and meteorological variables are provided on an hourly basis from the EMEP/MSW model. These EMEP model values are for a height of ca. 45 m, and essentially represent the influence of incoming advected air above the forest canopy. Concentrations of longer lived pollutants, including CO, NO_x and O₃ at 45 m and above are updated from the EMEP model

every time-step, whereas concentrations of BVOC and short-lived pollutants (OH, NO₃, etc.) are completely governed by ESX dispersion and chemistry.

In order to assess differences in the impact of BVOC and soil-NO on in-canopy O₃ and POD estimates across Europe, we have performed a number of runs of the EMEP-ESX combination in different regions, with the EMEP model providing location-specific meteorology and boundary concentrations, and ESX used for the fine-scale vertical modelling. We have performed these calculations for a full month, July 2012. This period has high photochemical activity at all sites.

To achieve comparability, we have made use of a generic forest class in all regions, and varied the emissions factors for isoprene, terpenes (mono- and sesqui-), and soil NO. This generic forest is set to have a height of 20.1 m, with ESX using 11 layers, each 2 m thick, for the canopy, extended with 30 more layers to a height of 1 km. The choice of canopy height was made so that the top 10 cm of the canopy corresponds to the 'upper-leaf' calculation required by the definition of POD as given by the UN-ECE Mapping Manual (LRTAP 2009, Mills et al. 2011b)

Emissions of BVOC are usually calculated by assigning standard emission factors to each species, where 'standard' emissions refer to those expected in full sunlight and at 30°C. In the EMEP system these emission factors are specified as micro-grammes emitted compounds per gramme foliar biomass and hour ($\mu\text{g g}^{-1} \text{h}^{-1}$), but such factors are readily converted to emissions per m² ground area and hour. Table 1 presents a summary of these area-based emission factors for some typical forest species in Europe. Base emission factors vary widely even within the same genus, e.g. Holm oak emits almost no isoprene but large amounts of monoterpenes, whereas the common Pendunculate and Downy oaks are large isoprene emitters but emit almost no terpenes.

Based upon these emission factors, we have explored the impact of letting isoprene standard emission factors vary from zero to 25 mg m⁻² h⁻¹ and terpene standard emission factors vary from zero to 4 mg m⁻² h⁻¹. Of course, environmental factors substantially modify such standard rates, and actual emissions will be much higher in regions with high temperatures.

In standard EMEP model usage, terpene emissions are handled with the surrogate α -pinene, but here we added two more reactive compounds: limonene, and β -caryophyllene. Especially β -caryophyllene, a sesqui-terpene, is far more reactive than α -pinene (Table 2), and hence more relevant for these in-canopy studies. Terpene emissions are assumed to consist of 60% α -pinene, 30% limonene, 10% β -caryophyllene. (β -caryophyllene has in fact been used in the EMEP model in other ECLAIRE research on the impacts of biotic stress on organic aerosol production from forests, see Bergström et al. (2014)).

Emissions of NO from soils are more difficult to define, in that they are typically rather sporadic and very sensitive to soil wetting and other events (e.g. Yienger and Levy 1995, Kesik et al. 2005, Butterbach-Bahl et al. 2009, Schaufler et al. 2010). Instead of

Table 1: Forest-specific biomass density and emission factors (leaf-level).

Species	Common name		Biomass density (D) g m^2	Emission factors ($\text{mg m}^{-2} \text{h}^{-1}$)	
				Isoprene	Terpenes
Betula	Birch		320	0	0.06
Carpinus bet.	European	Horn-beam	320	0	0.22
Quercus ilex.	Holm (Holly) Oak		500	0.05	15
Quercus petr.	Sessile oak		320	14.4	0.16
Quercus pube.	Downy oak		320	25.6	0.06
Quercus robu.	Pendunculate		320	0.01	0.1
Pinus pinaster	Maritime pine		700	0	1.05
Pinus pinea	Stone pine		700	0	4.2
Pinus sylvestris	Scots pine		700	0.07	2.1
Picea abies	Norway spruce		1400	1.4	2.1
Picea sitch.	Sitka spruce		1400	7.0	4.2

Notes: Biomass densities and emission factors from EMEP MSC-W model system (Simpson et al. 2012). Emission factors are at ‘standard’ conditions: full sunlight, 30°C.

calculating hourly variations, we have here tested the impact of either zero emissions, or emissions of $100 \text{ ng(NO)}/\text{m}^2/\text{s}$. The value of $100 \text{ ng(NO)}/\text{m}^2/\text{s}$ is comparable to some peak levels observed during the Bosco-Fontana campaign.

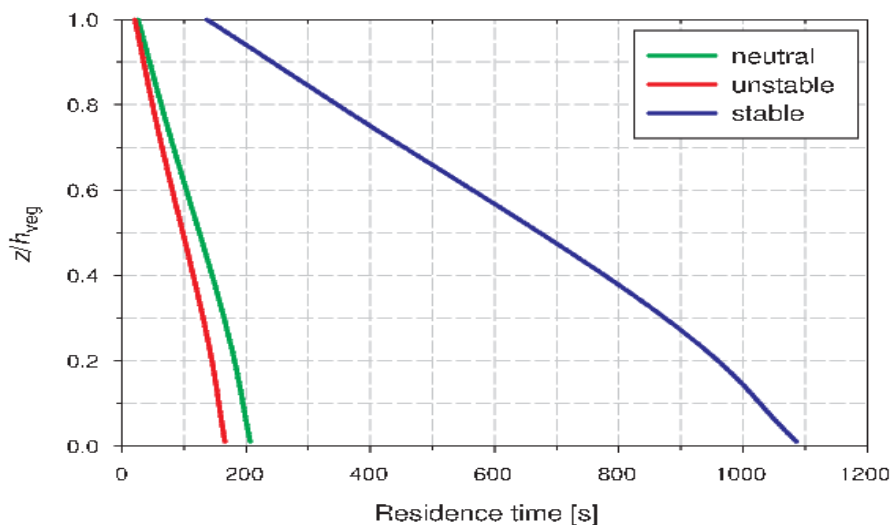


Figure 1: In-canopy residence time, here defined as the time after which less than $1/e$ of the initial mass at a certain height remains within the canopy. Three stability conditions are defined in terms of friction velocity (u^*) and Obukhov length (L): neutral, $u^* = 0.5 \text{ m s}^{-1}$, $1/L = 0$; unstable, $u^* = 0.2 \text{ m s}^{-1}$, $1/L = -0.05 \text{ m}^{-1}$; stable, $u^* = 0.2 \text{ m s}^{-1}$, $1/L = 0.02 \text{ m}^{-1}$. Vegetation height $h_{veg} = 10 \text{ m}$.

3 Results

As an example of the impacts of forests on mixing time-scales, Fig. 1 demonstrates how the residence time, which reflects the intensity of vertical mixing, varies within the vegetation canopy. In near-neutral conditions within a 10-m tall vegetation, this time ranges from about 3 minutes in the lower part to less than 0.5 min in the upper part of the canopy. Stable stratification suppresses turbulent mixing and potentially increases residence times considerably. For comparison, Table 2 presents some typical lifetimes of the BVOC species which we use here for ESX. It is clear that for the typically abundant species, isoprene and α -pinene, the chemical time-scales are substantially longer than these mixing time-scales, but the time-scale for reaction between O_3 and β -caryophyllene is clearly competitive with mixing time-scales. Of course, even moderately-reactive BVOC can impact concentrations of ozone and radicals if the BVOC concentrations in the canopy space are sufficiently high.

Figure 2 illustrates the impact of the soil-NO emission tests for a generic forest located at the Bosco-Fontana location in northern Italy, driven by EMEP model boundary conditions. Here we illustrate the development of vertical profiles of pollutants for a 12 h period over a forest with high BVOC and either high or zero soil-NO emissions. These calculations show, for example, a strong effect of the (admittedly high) soil-NO

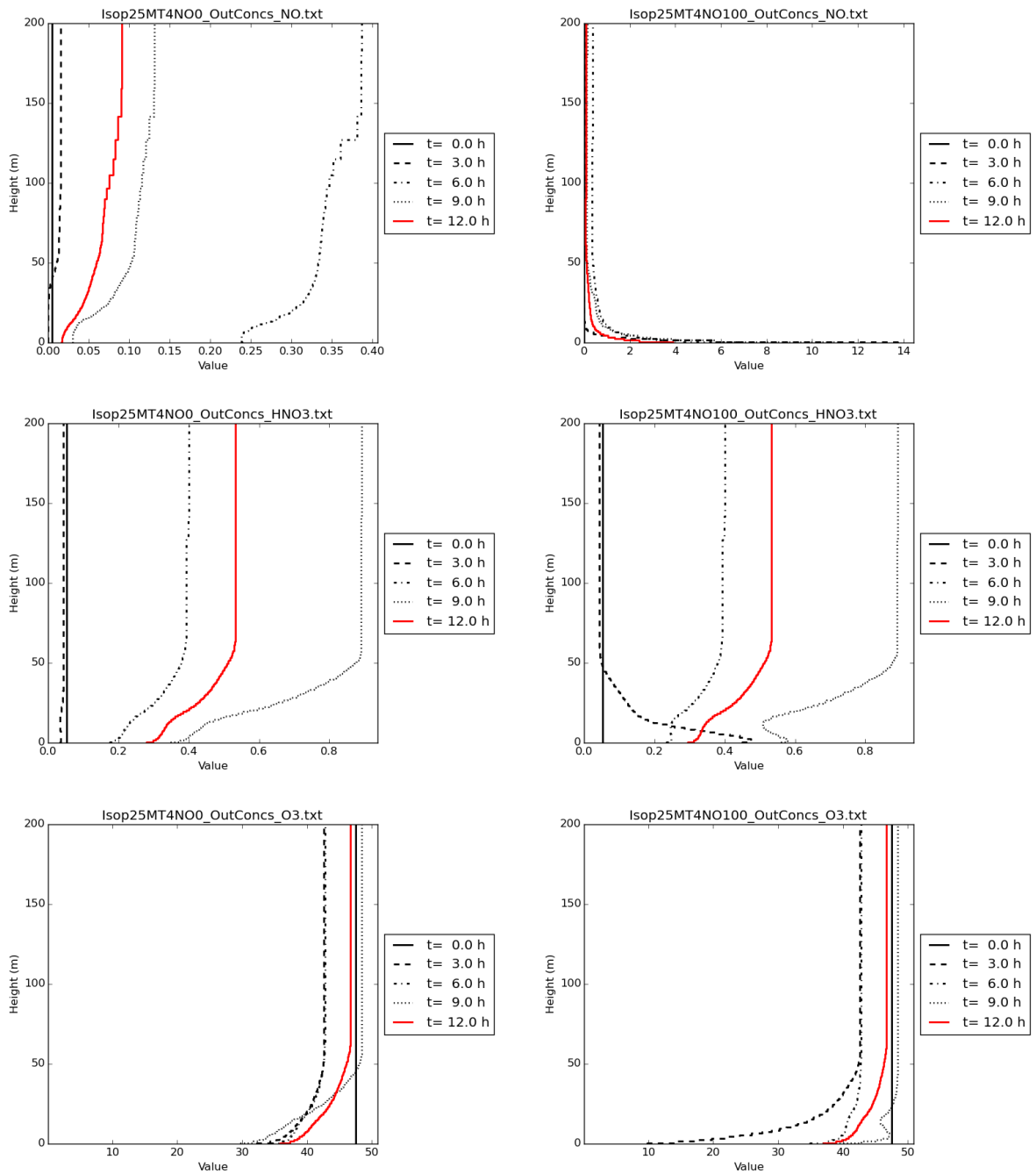


Figure 2: Examples of the impact of soil NO emissions on concentrations of NO, HNO₃ and O₃ in a forest canopy in northern Italy, as calculated with the ESX model. Concentrations are given from midnight (t=0 h) to mid-day (t=12 h), for a generic deciduous forest with high BVOC emissions. Results are given for NO (top row), HNO₃ (middle row), and O₃ (bottom-row). Results on left are for zero soil-NO emissions, results on right for high soil NO emissions (see text). All concentrations are in ppb. Note the large difference in x-axis scale between the NO plots.

Table 2: Estimated lifetimes of BVOC species used in ESX runs

Species	Lifetime (hours) due to reaction with		
	OH	O ₃	NO ₃
Isoprene	1.7	31	0.8
α -pinene	3.5	4.4	1.8
Limonene	1.1	2.0	0.9
β -caryophyllene	0.9	0.03	0.5

Notes: Assumes OH, O₃ and NO₃ concentrations of 1.5×10^6 , 7.0×10^{11} and 2.4×10^7 molecules cm⁻³, same as in Seinfeld and Pandis (1998)

emissions on the near-ground NO concentrations - much higher values than seen above. HNO₃, on the other hand, is changing at high elevation due to changes in the advected concentrations. Within the canopy, though, a mixture of processes is seen - deposition losses at most time-intervals but also HNO₃ production (due to conversion of soil-NO through NO₂ chemistry) causing concentration increases near the ground.

The impact of the isoprene and terpene emissions on the ozone deposition sinks was also examined, but found to be rather small. As an example, the left column of Fig. 3 shows the deposition loss terms for the different canopy layers. The upper plot assumes the highest emission rates for isoprene, terpenes and soil NO; the lower plot assumes zero emissions for these compounds. In this case, we present an example for a Spanish location (the site Miraflores), though still for our generic forest for comparability.

By eye, it is actually difficult to see much change in the ozone deposition terms seen in Fig. 3, except for the ground deposition which is somewhat greater with no biogenic emissions. This last effect is the simple result of removing the NO sink for ozone in the lowest layer.

Fig. 3 also illustrates the ozone fluxes per square meter of projected leaf area (PLA) (or of projected area for the non-leaf or ground-surface terms). These fluxes are clearly highest at the top of the canopy where solar radiation is maximised, also the region of highest wind-speed and hence lower resistances from the turbulence and diffusion processes. Further down the canopy radiation and wind-speed are reduced, as are ozone concentrations, and flux rates decrease.

As defined in the LRTAP Mapping Manual (LRTAP 2009, Mills et al. 2011b) the phyto-toxic ozone dose (POD) should be calculated for the very top of the canopy. With our setup of a 20.1 m high forest and 2 m layers in the canopy, this means that only stomatal fluxes in the top 10 cm of the canopy are relevant to the calculation of POD. There is thus an offset between the region of highest ozone losses (between ca. 14–18 m in Fig. 3, left column) and the region of highest flux values per m² leaf. In fact, the high ozone loss region has lower flux rates (right column) than seen at the top of canopy. As illustrated in the earlier examples, the biggest effects found with biogenic emissions

have usually been in the lowest canopy layers. Thus, from the examples presented so far, the preliminary finding is that top-of-canopy POD values are not very sensitive to in-canopy chemistry.

Figure 4 shows how the same soil-NO test as in Fig. 2 impacts the different deposition sinks of ozone for the generic forest located in Germany (same location as the Melpitz site), this time for a 31-day period (July 2012). Here we have separated the different deposition sinks. An interesting feature of these results is that although the stomatal sinks have the strongest diurnal cycle as expected, the various non-stomatal sinks also show strong diurnal variations. This reflects the impact of changes in turbulence and hence mixing through the day. Strong nighttime stability restricts for example exchange with the lowest model layer, and thus gives very low ground-surface deposition.

It is also apparent from Fig. 4 that the canopy-scale deposition fluxes are not affected to a large extent by these biogenic emissions considered here. The most obvious change is that the high emission case results in more frequent near-zero deposition fluxes at nighttime, and this can be attributed to soil-NO reacting with ozone in the lower canopy.

Table 3 presents the results for our generic forest at eight locations across Europe. Results are presented for the different components of the in-canopy deposition (ground deposition, stomatal and non-stomatal deposition to leaves, and to non-leaf surfaces (twigs, bark etc.). PODY results are also presented, with threshold fluxes of $Y = 0, 1$ and $2 \text{ nmole m}^{-2}(\text{PLA}) \text{ s}^{-1}$. As noted above, an important feature of these calculations is that the PODY values represent fluxes to the top of canopy, in our case the top 10 cm of the forest.

Some important features of these results can be summarised:

- The calculated PODY values are remarkably stable across these experiments. For example, at the Finnish site POD_1 values only vary between 1.40 to 1.49 $\text{mmole O}_3 \text{ m}^{-2} \text{ yr}^{-1}$ (a spread of ca. 5%). At the Italian location POD_1 values are lower, but with higher variability – between 1.15 to 1.28 (spread of 11%).
- The changes in total stomatal uptake to the canopy are far more stable. Our example Finnish site has total stomatal uptake varying between 14.4 to 14.9 $\text{mmole O}_3 \text{ m}^{-2} \text{ yr}^{-1}$ (a spread of only 3%). At the northern Italian location the equivalent values are 17.3 to 18.4 (6%).
- Larger changes are seen in the other canopy deposition terms, especially the ground deposition (e.g. Finland has 6.2 to 7.2 $\text{mmole O}_3 \text{ m}^{-2} \text{ yr}^{-1}$, spread 4%, Italy has 9.8 to 11.1 $\text{mmole O}_3 \text{ m}^{-2} \text{ yr}^{-1}$, spread 13%).
- Total deposition controls the lifetime of ozone, and this also changes significantly, e.g. Finland (6%), Spain (7%) or 8% for Italy.

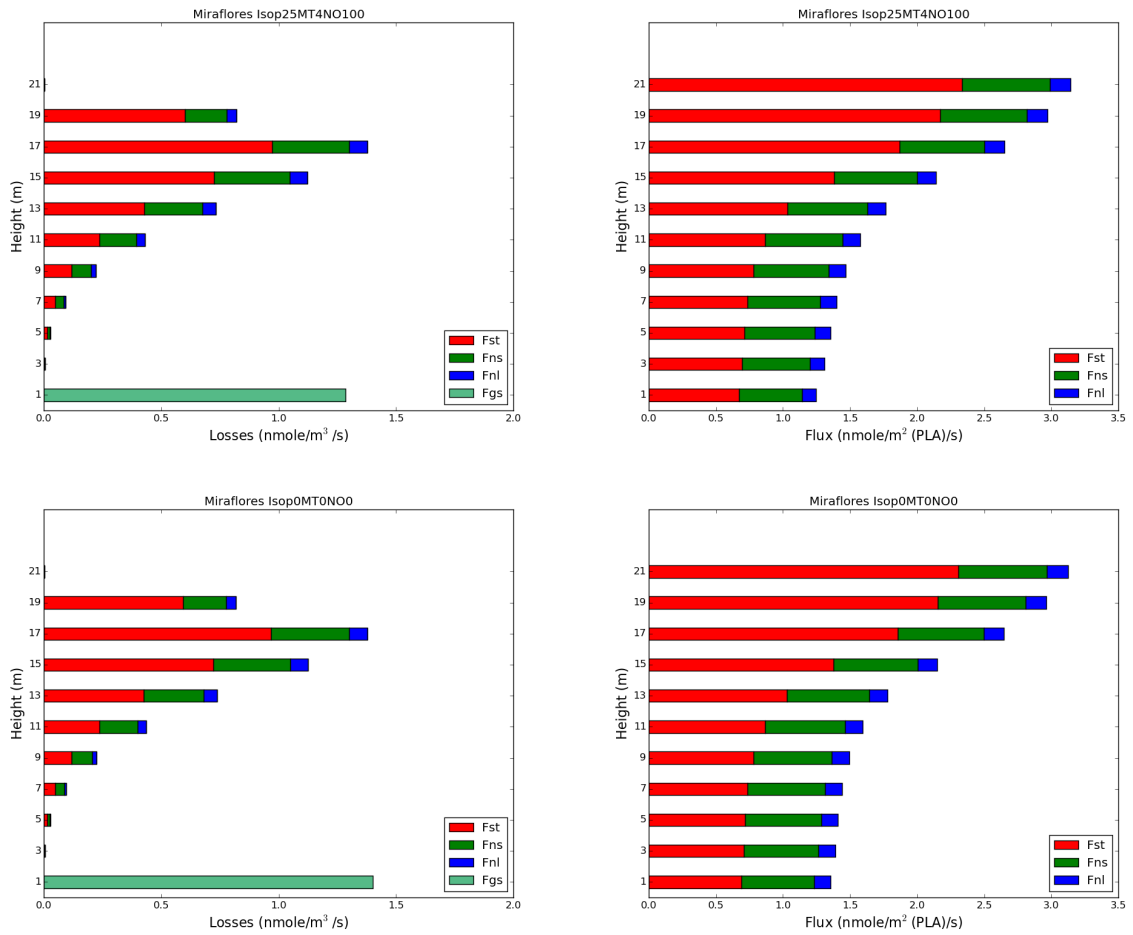


Figure 3: Ozone sink terms (left) and fluxes (right) for generic forest in Spain, with high BVOC and soil-NO emissions (top) and zero BVOC and soil-NO emissions (bottom). Terms account for stomatal fluxes (Fst), non-stomatal leaf fluxes (Fns), non-leaf fluxes (Fnl) and ground surface (Fgs). See text.

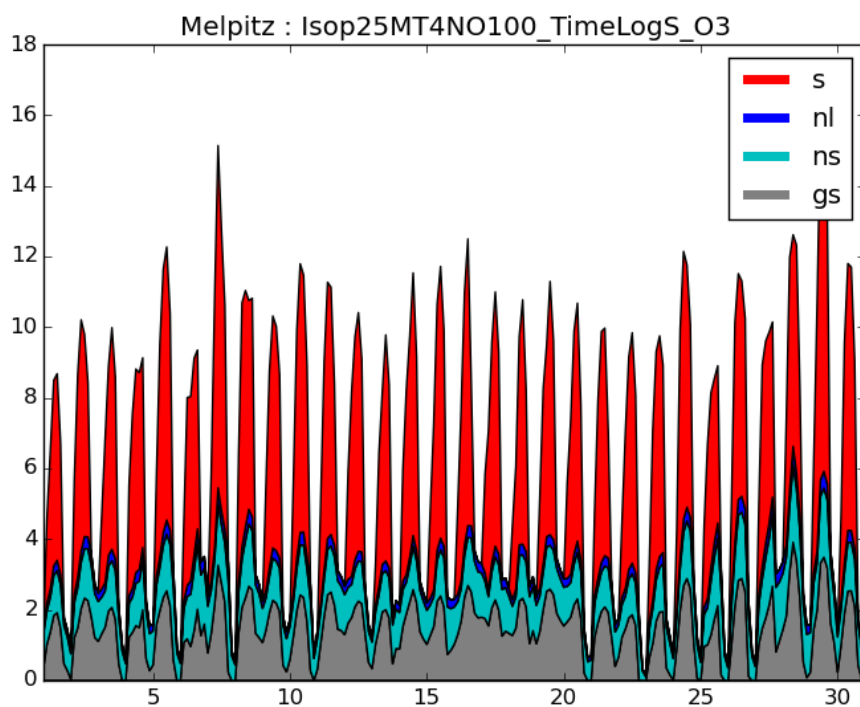
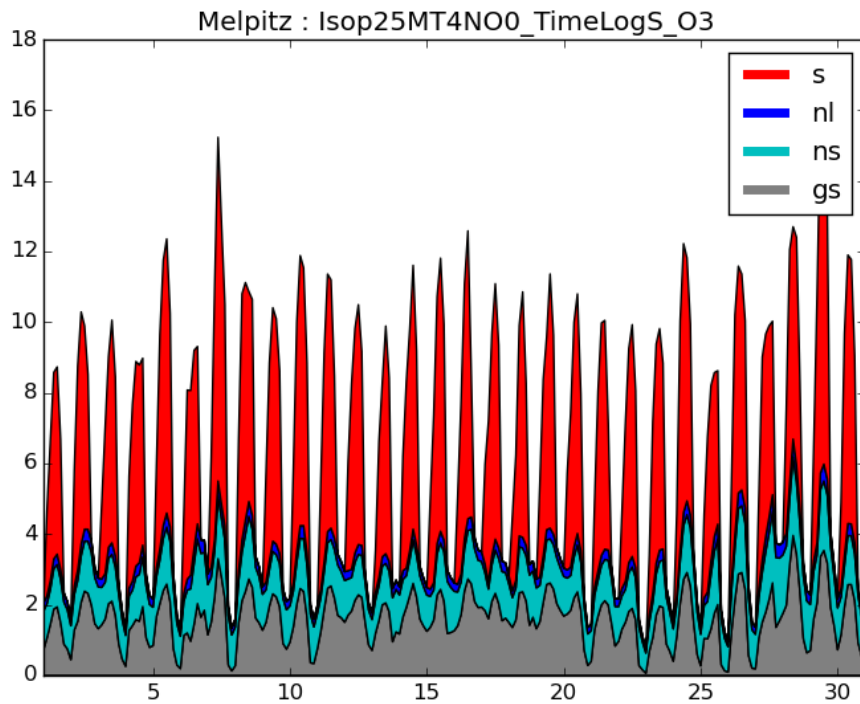


Figure 4: Impact of soil NO emissions on ozone deposition fluxes ($mmole\ O_3\ m^{-2}\ yr^{-1}$) for a forest canopy in Germany, as calculated with the EMEP-ESX model system. We here present results for a forest with high BVOC emissions. Results are given with zero (top) or high (bottom) soil-NO emissions. The sink terms are stomatal (s), non-leaf (nl), non-stomatal leaf (ns) and ground surface (gs).

- The biggest changes seem to arise from the soil-NO emission changes, and to a lesser extent isoprene changes.

The results presented above suggest that current estimates of BVOC and NO emissions have some impact on calculated POD estimates, but not too dramatic. One reason is that POD is based upon top-of-canopy O₃ flux estimates, whereas most canopy-chemistry interactions (especially with NO emitted from the forest floor) occur lower down, where time-scales are longer. Another reason is that most of the BVOC emissions are simply too unreactive to affect canopy O₃ levels to a significant extent. Soil-NO will react quickly with ozone, but in this case emission levels are again too low to affect within-canopy (and especially top of canopy) O₃ concentrations in a significant way.

These results also confirm the findings of the earlier EMEP CTM studies that stomatal fluxes are rather similar across Europe (Simpson et al. 2007, Mills et al. 2011a), even though O₃ concentrations (and concentration based metrics such as AOT40) vary significantly.

Table 3: Impact of varying isoprene (Eiso), terpene (Emt) and soil-NO (Eno) emissions on ozone deposition terms (mmole(O₃)/m² (ground area)/yr) and PODY values for July 2012. Calculations for generic forest in different locations, driven by EMEP CTM boundary conditions. Eiso and Emt are ‘standard’ emission factors for full sunlight, 30°C, in $\mu\text{g g}^{-1} \text{h}^{-1}$; Eno represents fixed emissions with units of ng(NO)/m²/s - see Sect. 2 for further details. POD has units mmole (O₃)/m² (PLA)/yr. The deposition terms are stomatal (s), non-leaf (nl), non-stomatal leaf (ns) and ground surface (gs).

Site	Emissions			Deposition terms					PODY (1 month)		
	Eiso	Emt	Eno	gs	s	ns	nl	Total	POD0	POD1	POD2
Hyytiala	25	0	0	7.170	14.700	5.570	1.280	28.720	2.05	1.42	0.82
Hyytiala	25	4	0	7.160	14.700	5.550	1.270	28.680	2.05	1.42	0.82
Hyytiala	10	0	0	7.170	14.700	5.570	1.280	28.720	2.05	1.42	0.82
Hyytiala	10	4	0	7.160	14.700	5.560	1.270	28.690	2.05	1.42	0.82
Hyytiala	0	0	0	7.150	14.700	5.560	1.280	28.690	2.05	1.42	0.81
Hyytiala	0	4	0	7.150	14.700	5.560	1.270	28.680	2.05	1.42	0.81
Hyytiala	25	0	100	6.730	14.800	5.650	1.290	28.470	2.11	1.48	0.87
Hyytiala	25	4	100	6.830	14.800	5.700	1.300	28.630	2.12	1.49	0.88
Hyytiala	10	0	100	6.380	14.600	5.420	1.240	27.640	2.07	1.44	0.84
Hyytiala	10	4	100	6.450	14.700	5.470	1.260	27.880	2.08	1.45	0.85
Hyytiala	0	0	100	6.210	14.400	5.220	1.200	27.030	2.02	1.40	0.79
Hyytiala	0	4	100	6.270	14.500	5.280	1.210	27.260	2.04	1.41	0.80
AuchencorthMoss	25	0	0	7.600	12.600	5.410	1.230	26.840	1.95	1.37	0.80
AuchencorthMoss	25	4	0	7.620	12.600	5.420	1.230	26.870	1.96	1.37	0.81
AuchencorthMoss	10	0	0	7.560	12.500	5.390	1.220	26.670	1.95	1.36	0.80
AuchencorthMoss	10	4	0	7.580	12.600	5.400	1.220	26.800	1.95	1.37	0.80
AuchencorthMoss	0	0	0	7.520	12.500	5.370	1.220	26.610	1.94	1.36	0.79
AuchencorthMoss	0	4	0	7.540	12.500	5.380	1.220	26.640	1.95	1.36	0.80
AuchencorthMoss	25	0	100	6.810	12.400	5.180	1.180	25.570	1.93	1.35	0.78
AuchencorthMoss	25	4	100	6.840	12.400	5.200	1.180	25.620	1.94	1.35	0.79
AuchencorthMoss	10	0	100	6.750	12.400	5.150	1.170	25.470	1.92	1.34	0.78
AuchencorthMoss	10	4	100	6.780	12.400	5.170	1.180	25.530	1.93	1.34	0.78
AuchencorthMoss	0	0	100	6.710	12.300	5.130	1.170	25.310	1.92	1.33	0.77
AuchencorthMoss	0	4	100	6.740	12.300	5.150	1.170	25.360	1.92	1.34	0.77

Continued on next page

Table 3 – continued from previous page

Site	Eiso	Emt	Eno	gs	s	ns	nl	Total	POD0	POD1	POD2
Melpitz	25	0	0	8.690	17.200	6.860	1.570	34.320	2.30	1.74	1.19
Melpitz	25	4	0	8.680	17.200	6.830	1.560	34.270	2.30	1.74	1.19
Melpitz	10	0	0	8.650	17.200	6.840	1.560	34.250	2.30	1.73	1.18
Melpitz	10	4	0	8.650	17.200	6.810	1.560	34.220	2.30	1.73	1.18
Melpitz	0	0	0	8.590	17.100	6.800	1.550	34.040	2.29	1.72	1.17
Melpitz	0	4	0	8.590	17.100	6.780	1.550	34.020	2.29	1.73	1.17
Melpitz	25	0	100	7.890	17.100	6.600	1.510	33.100	2.31	1.74	1.19
Melpitz	25	4	100	7.950	17.100	6.630	1.520	33.200	2.31	1.75	1.20
Melpitz	10	0	100	7.670	17.000	6.460	1.480	32.610	2.29	1.72	1.17
Melpitz	10	4	100	7.730	17.000	6.490	1.490	32.710	2.29	1.73	1.18
Melpitz	0	0	100	7.520	16.900	6.330	1.450	32.200	2.27	1.70	1.15
Melpitz	0	4	100	7.580	16.900	6.370	1.460	32.310	2.27	1.71	1.16
Cabauw	25	0	0	7.610	15.600	6.010	1.380	30.600	2.30	1.74	1.19
Cabauw	25	4	0	7.630	15.600	6.000	1.380	30.610	2.31	1.75	1.20
Cabauw	10	0	0	7.490	15.500	5.920	1.360	30.270	2.28	1.72	1.17
Cabauw	10	4	0	7.520	15.500	5.920	1.360	30.300	2.29	1.73	1.18
Cabauw	0	0	0	7.360	15.400	5.800	1.330	29.890	2.25	1.69	1.13
Cabauw	0	4	0	7.390	15.400	5.810	1.330	29.930	2.26	1.70	1.14
Cabauw	25	0	100	6.770	15.400	5.770	1.330	29.270	2.30	1.74	1.19
Cabauw	25	4	100	6.830	15.500	5.800	1.330	29.460	2.31	1.75	1.20
Cabauw	10	0	100	6.560	15.300	5.570	1.280	28.710	2.26	1.71	1.15
Cabauw	10	4	100	6.600	15.300	5.610	1.290	28.800	2.28	1.72	1.16
Cabauw	0	0	100	6.420	15.200	5.340	1.230	28.190	2.22	1.66	1.11
Cabauw	0	4	100	6.460	15.200	5.390	1.250	28.300	2.23	1.67	1.12
Grignon	25	0	0	7.800	15.600	5.960	1.360	30.720	1.98	1.42	0.88
Grignon	25	4	0	7.840	15.700	5.970	1.360	30.870	1.99	1.44	0.89
Grignon	10	0	0	7.670	15.500	5.870	1.340	30.380	1.95	1.40	0.86
Grignon	10	4	0	7.710	15.600	5.880	1.350	30.540	1.96	1.41	0.87
Grignon	0	0	0	7.550	15.400	5.780	1.320	30.050	1.93	1.37	0.84
Grignon	0	4	0	7.580	15.500	5.790	1.330	30.200	1.94	1.38	0.84
Grignon	25	0	100	6.900	15.400	5.640	1.290	29.230	1.96	1.41	0.87
Grignon	25	4	100	6.960	15.500	5.680	1.300	29.440	1.97	1.42	0.88
Grignon	10	0	100	6.690	15.300	5.500	1.270	28.760	1.93	1.38	0.84
Grignon	10	4	100	6.750	15.400	5.540	1.270	28.960	1.94	1.39	0.85
Grignon	0	0	100	6.530	15.200	5.360	1.240	28.330	1.90	1.35	0.82
Payerne	25	0	0	8.120	17.300	6.750	1.550	33.720	2.22	1.68	1.14
Payerne	25	4	0	8.100	17.400	6.720	1.540	33.760	2.22	1.68	1.14
Payerne	10	0	0	8.090	17.300	6.730	1.540	33.660	2.21	1.67	1.14
Payerne	10	4	0	8.080	17.300	6.710	1.540	33.630	2.22	1.67	1.14
Payerne	0	0	0	8.040	17.300	6.700	1.540	33.580	2.21	1.66	1.13
Payerne	0	4	0	8.040	17.300	6.680	1.530	33.550	2.21	1.67	1.13
Payerne	25	0	100	7.450	17.300	6.570	1.510	32.830	2.23	1.69	1.15
Payerne	25	4	100	7.500	17.300	6.590	1.510	32.900	2.23	1.69	1.16
Payerne	10	0	100	7.220	17.200	6.420	1.480	32.320	2.21	1.67	1.13
Payerne	10	4	100	7.260	17.300	6.450	1.480	32.490	2.22	1.68	1.14
Payerne	0	0	100	7.080	17.100	6.290	1.450	31.920	2.19	1.65	1.11
Payerne	0	4	100	7.120	17.200	6.320	1.460	32.100	2.20	1.65	1.12
BoscoFontana	25	0	0	11.100	18.000	8.730	1.990	39.820	1.74	1.21	0.69
BoscoFontana	25	4	0	11.100	17.900	8.710	1.980	39.690	1.75	1.21	0.69
BoscoFontana	10	0	0	11.100	17.900	8.740	1.990	39.730	1.74	1.20	0.68
BoscoFontana	10	4	0	11.100	17.900	8.720	1.980	39.700	1.74	1.21	0.68
BoscoFontana	0	0	0	10.900	17.700	8.660	1.970	39.230	1.71	1.18	0.65
BoscoFontana	0	4	0	11.000	17.800	8.670	1.970	39.440	1.72	1.19	0.66
BoscoFontana	25	0	100	10.900	18.200	8.840	2.010	39.950	1.79	1.26	0.73

Continued on next page

Table 3 – continued from previous page

Site	Eiso	Emt	Eno	gs	s	ns	nl	Total	POD0	POD1	POD2
BoscoFontana	25	4	100	11.100	18.400	8.890	2.020	40.410	1.81	1.28	0.75
BoscoFontana	10	0	100	10.400	17.900	8.680	1.970	38.950	1.75	1.22	0.69
BoscoFontana	10	4	100	10.700	18.100	8.750	1.990	39.540	1.78	1.24	0.72
BoscoFontana	0	0	100	9.790	17.300	8.400	1.910	37.400	1.68	1.15	0.62
BoscoFontana	0	4	100	9.950	17.500	8.470	1.930	37.850	1.70	1.17	0.64
Miraflores	25	0	0	7.580	16.900	7.550	1.730	33.760	2.12	1.60	1.08
Miraflores	25	4	0	7.580	16.900	7.540	1.730	33.750	2.12	1.60	1.09
Miraflores	10	0	0	7.550	16.800	7.520	1.720	33.590	2.11	1.59	1.08
Miraflores	10	4	0	7.550	16.800	7.520	1.720	33.590	2.12	1.60	1.08
Miraflores	0	0	0	7.480	16.700	7.480	1.710	33.370	2.10	1.58	1.07
Miraflores	0	4	0	7.490	16.800	7.480	1.710	33.480	2.11	1.59	1.07
Miraflores	25	0	100	6.790	16.700	7.280	1.670	32.440	2.11	1.59	1.08
Miraflores	25	4	100	6.860	16.800	7.320	1.680	32.660	2.12	1.60	1.09
Miraflores	10	0	100	6.600	16.600	7.190	1.650	32.040	2.10	1.58	1.06
Miraflores	10	4	100	6.670	16.700	7.230	1.660	32.260	2.10	1.59	1.07
Miraflores	0	0	100	6.420	16.400	7.100	1.630	31.550	2.08	1.56	1.04
Miraflores	0	4	100	6.490	16.500	7.140	1.640	31.770	2.08	1.57	1.05

3.1 Remarks

The results presented above are the first of their kind with the newly developed EMEP-ESX combination. Once the full data analysis of the ECLAIRE C1 sites is available (including allocation of losses to different sinks), the process of evaluating ESX on a site-by-site basis can begin, and the model can be evaluated and adapted to local conditions as appropriate.

Thus, the main need in the near future is to compare the ESX model with these field measurements. Indeed, a large amount of work will be needed to thoroughly test the ESX system, and to gain experience of its properties. All 1-D models show great sensitivity to input parameters and assumptions, and thus such models can never be fully validated. However, our aim is to have a system which captures the main features of ecosystem-atmosphere exchange and helps to improve the modelling of such exchanges in future EMEP assessments.

It can be noted though that all such calculations of the impacts of BVOC and soil-NO are fraught with uncertainty. Many problems have been identified in both the emission rates (including those of unknown compounds) and chemical mechanisms involved in BVOC photo-chemistry, for example (Makar et al. 1999, Stroud et al. 2005, Mogensen et al. 2011, Noelscher et al. 2012, Wolfe et al. 2011, Whalley et al. 2014, Squire et al. 2015). For soil-NO emissions, a deterministic estimate would require knowledge of soil microbiology and pH as well as wetness status (Yienger and Levy 1995, Ludwig et al. 2001, Kesik et al. 2005, Butterbach-Bahl et al. 2009, Schindlbacher et al. 2004).

Given these uncertainties, our preliminary analysis is re-assuring in the sense that the top-of-canopy metrics such as POD are rather insensitive to the in-canopy reactions. The impact on total O₃ losses is more significant, and this will be a main focus of future

activities.

4 Milestones achieved

MS28 Implementation and initial testing of coupled model system

5 Deviations and reasons

The decision was made early in ECLAIRE to construct a new 1-dimensional model, rather than to adapt an existing model. The new model, ESX, has taken considerable resources and time, and full coupling within EMEP was not possible within the project. Still, the combination of EMEP and ESX as used here is likely an even more powerful tool than originally envisaged.

6 Publications

Bergström, R.B., Carbonaceous Aerosol in Europe. Out of the woods and into the blue?, PhD Thesis, Dept. Chemistry & Molec. Biology, University of Gothenburg, Sweden, ISBN 978-91-628- 9505-1, Sept. 2015

Bergström, R., Hallquist, M., Simpson, D., Wildt, J. & Mentel, T. F. Biotic stress: a significant contributor to organic aerosol in Europe? *Atmospheric Chemistry and Physics*, 2014, 14, 13643-13660

Cieslik, S., Tuovinen, J.-P., Baumgarten, M., Matyssek, R., Brito, P. and Wieser, G., 2013. Gaseous exchange between forests and the atmosphere. *Developments in Environmental Science* 13, 19- 36.

Simpson, D. and Tuovinen, J.-P., 2014. ECLAIRE Ecosystem Surface Exchange model (ESX). In: *Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. EMEP Status report 1/2014*, Norwegian Meteorological Institute, 147-154, 2014.

Simpson, D. and Tuovinen, J.-P., Towards a flexible 1-D modelling system for biosphere-atmosphere exchange: the Ecosystem Surface Exchange (ESX) model, 2015 in preparation

7 Meetings

Participation in ECLAIRE annual meetings, plus:

- 19-21.03.2012, Edinburgh,
ECLAIRE meeting on multi-layer model harmonisation.

- 20/5-10/6 2013
Visit of Alan Briolat (SEI-Y) to David Simpson (MET Norway, Chalmers) to work on ESX and DO3SE codes.
- 28-29.09.2013, Amsterdam,
Work-meeting for ECLAIRE ESX model development
- 18-19/11/2013, Oslo, internal meeting
EMEP informal group meeting on model development. Presentation and discussion of the ECLAIRE ESX work.
- 23/9-11/10 2013
Visit of Alan Briolat (SEI-Y) to David Simpson (MET Norway, Chalmers) to work on ESX and DO3SE codes.
- 2-6/02/2014, Paris,
ECLAIRE Winter school on biosphere-atmosphere exchange. David Simpson was one of the teachers and presented both ESX and EMEP work.
- 19-22/5/2014, Edinburgh,
Meeting with CEH and colleagues from Garmisch (KIT) to discuss approaches to link ecosystem modelling with EMEP/ESX. Focus on Landscape DNDC model.
- 25-28/5/2014, Edinburgh,
Meeting with ECLAIRE ESX+DEWS development group, CEH and University of Manchester colleagues
- 22/6-4/7 2014, Gothenburg,
Visit of Juha-Pekka Tuovinen (FMI) to David Simpson (MET Norway, Chalmers) to work on ESX.
- 26-30.01.2015, Gothenburg
Visit of Matthias Karl (NILU) to David Simpson (MET Norway, Chalmers) to implement aerosol dynamics code (MAFOR) in ESX and EMEP.
- 11-13.05.2015, Gothenburg
Visit of Raia Massad (INRA) to David Simpson (MET Norway, Chalmers) to work on ESX/EMEP and N-exchange.
- 9-21.03.2015, Gothenburg,
Visit of Juha-Pekka Tuovinen (FMI) to David Simpson (MET Norway, Chalmers) to work on ESX.

8 List of Documents/Annexes:

n.a.

References

- Bergström, R., Hallquist, M., Simpson, D., Wildt, J., and Mentel, T. F.: Biotic stress: a significant contributor to organic aerosol in Europe?, *Atmospheric Chemistry and Physics*, 14, 13 643–13 660, doi:10.5194/acp-14-13643-2014, URL <http://www.atmos-chem-phys.net/14/13643/2014/>, 2014.
- Butterbach-Bahl, K., Kahl, M., Mykhayliv, L., Werner, C., Kiese, R., and Li, C.: A European-wide inventory of soil NO emissions using the biogeochemical models DNDC/Forest-DNDC, *Atmos. Environ.*, 43, 1392 – 1402, doi:10.1016/j.atmosenv.2008.02.008, 2009.
- Kesik, M., Ambus, P., Baritz, R., Brüggemann, N., Butterbach-Bahl, K., Damm, M., Duyzer, J., Horváth, L., Kiese, R., Kitzler, B., Leip, A., Li, C., Pihlatie, M., Pilegaard, K., Seufert, G., Simpson, D., Skiba, U., Smiatek, G., Vesala, T., and Zechmeister-Boltenstern, S.: Inventory of N₂O and NO emissions from European forest soils, *Biogeosciences*, 2, 353–375, 2005.
- LRTAP: Mapping critical levels for vegetation, in: *Manual on Methodologies and Criteria for Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends. Revision of 2009*, edited by Mills, G., UNECE Convention on Long-range Transboundary Air Pollution. International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops, updated version available at www.icpmapping.com/, 2009.
- Ludwig, J., Meixner, F., Vogel, B., and Förstner, J.: Soil-air exchange of nitric oxide: An overview of processes, environmental factors, and modeling studies, *Biogeochem.*, 52, 225–257, 2001.
- Makar, P., Fuentes, J., Wang, D., Staebler, R., and Wiebe, H.: Chemical processing of biogenic hydrocarbons within and above a temperate deciduous forest, *J. Geophys. Res.*, 104, 3581–3603, 1999.
- Mills, G., Hayes, F., Simpson, D., Emberson, L., Norris, D., Harmens, H., and Büker, P.: Evidence of widespread effects of ozone on crops and (semi-) natural vegetation in Europe (1990-2006) in relation to AOT40- and flux-based risk maps, *Global Change Biology*, 17, 592–613, doi:10.1111/j.1365-2486.2010.02217.x, URL <http://dx.doi.org/10.1111/j.1365-2486.2010.02217.x>, 2011a.

- Mills, G., Pleijel, H., Braun, S., Büker, P., Bermejo, V., Calvo, E., Danielsson, H., Emberson, L., Grünhage, L., Fernández, I. G., Harmens, H., Hayes, F., Karlsson, P.-E., and Simpson, D.: New stomatal flux-based critical levels for ozone effects on vegetation, *Atmos. Environ.*, 45, 5064 – 5068, doi:10.1016/j.atmosenv.2011.06.009, 2011b.
- Mogensen, D., Smolander, S., Sogachev, A., Zhou, L., Sinha, V., Guenther, A., Williams, J., Nieminen, T., Kajos, M. K., Rinne, J., Kulmala, M., and Boy, M.: Modelling atmospheric OH-reactivity in a boreal forest ecosystem, *Atmos. Chem. Physics*, 11, 9709–9719, doi:10.5194/acp-11-9709-2011, 2011.
- Noelscher, A. C., Williams, J., Sinha, V., Custer, T., Song, W., Johnson, A. M., Axinte, R., Bozem, H., Fischer, H., Pouvesle, N., Phillips, G., Crowley, J. N., Rantala, P., Rinne, J., Kulmala, M., Gonzales, D., Valverde-Canossa, J., Vogel, A., Hoffmann, T., Ouwersloot, H. G., de Arellano, J. V.-G., and Lelieveld, J.: Summertime total OH reactivity measurements from boreal forest during HUMPPA-COPEC 2010, *Atmospheric Chemistry and Physics*, 12, 8257–8270, doi:10.5194/acp-12-8257-2012, 2012.
- Schaufler, G., Kitzler, B., Schindlbacher, A., Skiba, U., Sutton, M. A., and Zechmeister-Boltenstern, S.: Greenhouse gas emissions from European soils under different land use: effects of soil moisture and temperature, *European Journal of Soil Science*, 61, 683–696, doi:10.1111/1.1365-2389.2010.01277.x, 2010.
- Schindlbacher, A., Zechmeister-Boltenstern, S., and Butterbach-Bahl, K.: Effects of soil moisture and temperature on NO, NO₂, and N₂O emissions from European forest soils, *J. Geophys. Res.*, 109, D17 302, doi:10.1029/2004JD004590, 2004.
- Seinfeld, J. and Pandis, S.: *Atmospheric chemistry and physics. From air pollution to climate change*, John Wiley and Sons, inc., New York, 1998.
- Simpson, D. and Tuovinen, J.-P.: ECLAIRE Ecosystem Surface Exchange model (ESX), in: *Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. Status Report 1/2014*, pp. 147–154, The Norwegian Meteorological Institute, Oslo, Norway, 2014.
- Simpson, D., Emberson, L., Ashmore, M., and Tuovinen, J.: A comparison of two different approaches for mapping potential ozone damage to vegetation. A model study, *Environ. Poll.*, 146, 715–725, doi:10.1016/j.envpol.2006.04.013, 2007.
- Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P.,

- Valdebenito, A., and Wind, P.: The EMEP MSC-W chemical transport model – technical description, *Atmos. Chem. Physics*, 12, 7825–7865, doi:10.5194/acp-12-7825-2012, URL <http://www.atmos-chem-phys.net/12/7825/2012/acp-12-7825-2012.html>, 2012.
- Squire, O. J., Archibald, A. T., Griffiths, P. T., Jenkin, M. E., Smith, D., and Pyle, J. A.: Influence of isoprene chemical mechanism on modelled changes in tropospheric ozone due to climate and land use over the 21st century, *Atmospheric Chemistry and Physics*, 15, 5123–5143, doi:10.5194/acp-15-5123-2015, URL <http://www.atmos-chem-phys.net/15/5123/2015/>, 2015.
- Stroud, C., Makar, P., Karl, T., Guenther, A., Geron, C., Turnipseed, A., Nemitz, E., Baker, B., Potosnak, M., and Fuentes, J.: Role of canopy-scale photochemistry in modifying biogenic-atmosphere exchange of reactive terpene species: Results from the CELTIC field study, *J. Geophys. Res.*, 110, doi:10.1029/2005JD005775, 2005.
- Whalley, L., Stone, D., and Heard, D.: New Insights into the Tropospheric Oxidation of Isoprene: Combining Field Measurements, Laboratory Studies, Chemical Modelling and Quantum Theory, *Atmospheric and Aerosol Chemistry*, 339, 55–95, doi:10.1007/128_2012_359, 2014.
- Wolfe, G. M., Thornton, J. A., McKay, M., and Goldstein, A. H.: Forest-atmosphere exchange of ozone: sensitivity to very reactive biogenic VOC emissions and implications for in-canopy photochemistry, *Atmos. Chem. Physics*, 11, 7875–7891, doi:10.5194/acp-11-7875-2011, 2011.
- Yienger, J. and Levy, H.: Empirical model of global soil-biogenic NO_x emissions, *J. Geophys. Res.*, 100, 11 447–11 464, 1995.