REPORT

Report on ship plume simulations and analysis

Deliverable D2.1.3.5 to project QUANTIFY

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R Report Summary

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Summary

The nonlinear chemistry of ozone formation from oxides of nitrogen causes that NOx emissions, that are in a regional-scale or a global models distributed directly over large grid squares, form more ozone than if they were treated in a gradually dispersing plume. An efficient method of plume parameterisation has been described by Cariolle et al. (2009) where the plume stage of emission is treated as a tracer and parameterised reaction rates for ozone and NOx losses in plume are given for aircraft plume. The rates of reactions affecting ozone, NOx and OH have been studied with a plume model of Lagrangean type and a loss term for NOx and OH in plume is defined for the parameterisation.

Contents

1 Introduction

Emission of exhaust gases and particles from seagoing ships contribute significantly to the anthropogenic emissions thereby affecting the chemical composition of the atmosphere, local and regional air quality and climate. The emitted compounds are injected into the atmosphere in form of coherent plumes, often in relatively pristine parts of the MBL. Models studying effects of these emissions are, however, using inventories that are calculated from emission totals distributed over the model domain with the help of spatial proxies of ship traffic. These emissions are instantaneously spread onto large inventory grid boxes. It is widely recognized that nonlinearities in atmospheric processes make the global-scale distributions sensitive to mixing processes. The plume processes thus may significantly affect the large scale distribution of chemical species in the atmosphere and effects of these subgrid-scale processes need to be accounted for.

2 Methodology

The chemical transformation of ship emissions in plumes were further investigated with a plume version of MOCCA model developed for this purpose during the Quantify project. The nonlinear chemistry of ozone formation from oxides of nitrogen causes that NOx emissions that are in a regional-scale or a global models distributed directly over large grid squares, form more ozone than if they were treated in a gradually dispersing plume. A method of plume parameterisation has been developed by Cariolle et al. (2009) where the plume stage of emission is treated as a NOx emission tracer. Parameterised first order reaction rates for ozone (Keff*[tracer]) and relative NOx loss (β) in plume are given for aircraft plume and Keff for ozone is also suggested for ship plumes in this study. The rates of reactions affecting ozone, NOx and OH were further investigated with a plume model. First the magnitude of the subscale effects was investigated by comparing an explicit-plume dispersion simulation (ED) with simulation where emission is instantly spread over a grid with size 25x25 km (ID). Second, the plume parameterisation as described by Cariolle et al. (2009) was implemented to the model and tested against the ED and ID simulations. Third, a new removal rate for the NOx tracer was added to this parameterisation to improve its performance.

The setup of simulations used in this study is described in Table 1. Cases with different photochemical activity and with different levels of background NOx were investigated.

The concept of the parameterisation is that the plume-stage NOx tracer has a constant lifetime τ. A situation of the ship emissions being released in a constant rate on a certain area was assumed in plume simulations. The Lagrangian perception of such situation is that the emission tracer is kept as a plume-stage tracer until time $t = \tau$ and then released as NOx. To avoid differences between ED, ID and IDP simulations caused by the fact that the maximum in plume chemical activity is shifted in time, and hence in photolytical activity in atmosphere due to the changing insulation, the simulations kept photolytical activity constant several hours after the plume release.

Parameters and initial concentrations used in plume model simulations were those of the 2007 Quantify campaign (Table 2). The model was initiated 36-40 hours prior to the ship emission with clean marine conditions. The plume mixing corresponds to stable cloud- free summer MBL with moderate winds.

Table 1. Setup of model simulations.

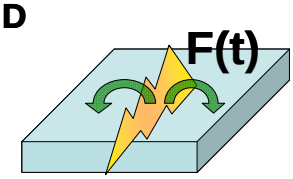
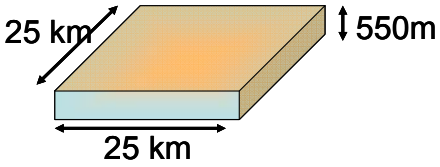
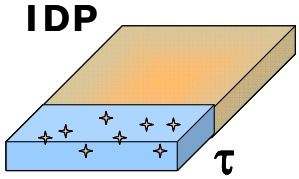
ED 	**Explicit plume dispersion simulation** using detailed time dependent dilution rate: Time dependent dilution rate F(t) arising from LES/Lagrangian plume simulation (Chosson et al., 2008) is used to describe plume/background mixing rate. The time evolution of F(t) is described by the function: $F(t) = b/tc$, t is time after the emission in minutes and b and c are fitting parameters.
ID 	**Instantaneous emission release simulation** - A common large scale model which consists in an instantaneous release of the chemical species at the beginning of the run period
IDP 	**Parameterised ID simulation** - A common large scale model where emission is released as a reactive tracer that is after the time τ converted into NOx. This simulation corresponds to modified parameterisation as described by Cariolle et al. (2009) for a constant emission

Table 2. Parameters b,c, of dispersion equation used in ED plume model simulations. Emissions of the simulated ships are given in g/s, in case of NOx molecular weight of NO was used.

b	1.659
c	1.133
CO_2 (g/s)	3111
NO_x (g/s)	66.9
SO_2 (g/s)	35.6

3 Results and discussion

The model simulations have shown strong nonlinearities in ozone formation in plumes emitted at daytime. In summer situation ozone formation through NO + HO_2 and NO + organic peroxy radicals, followed by the NO_2 photolysis, was in explicit plumes less than a half of ozone formed in simulations where emissions were directly spread over a 25x25 km grid (Figure 1a). Effect was similar for the OH concentrations (Figure 1b). The parameterisation as suggested in Cariole et al. (2008) have not changed the amount of ozone or OH formed in the plume, only shifted concentration evolution in time. This is an expected result as the Keff removing the plume-stage ozone is very low in the MBL and the plume-stage tracer behaves as a nonreactive tracer. This can be seen in Figure 1 where the dashed green line is simulation with non-reactive tracer and the green triangles are simulation with tracer affecting ozone with the suggested Keff. We need to remove some NOx in the parameterisation without forming ozone to obtain result corresponding to the ED case. The night-time emitted plumes exhibit nonlinear effects that are different from the

daytime: As there is no ozone formation during the night, reactions of NOx in the marine boundary layer, however, following daytime. The difference in the NOx sink betwee heterogeneous reactions on sea-salt with some contributio

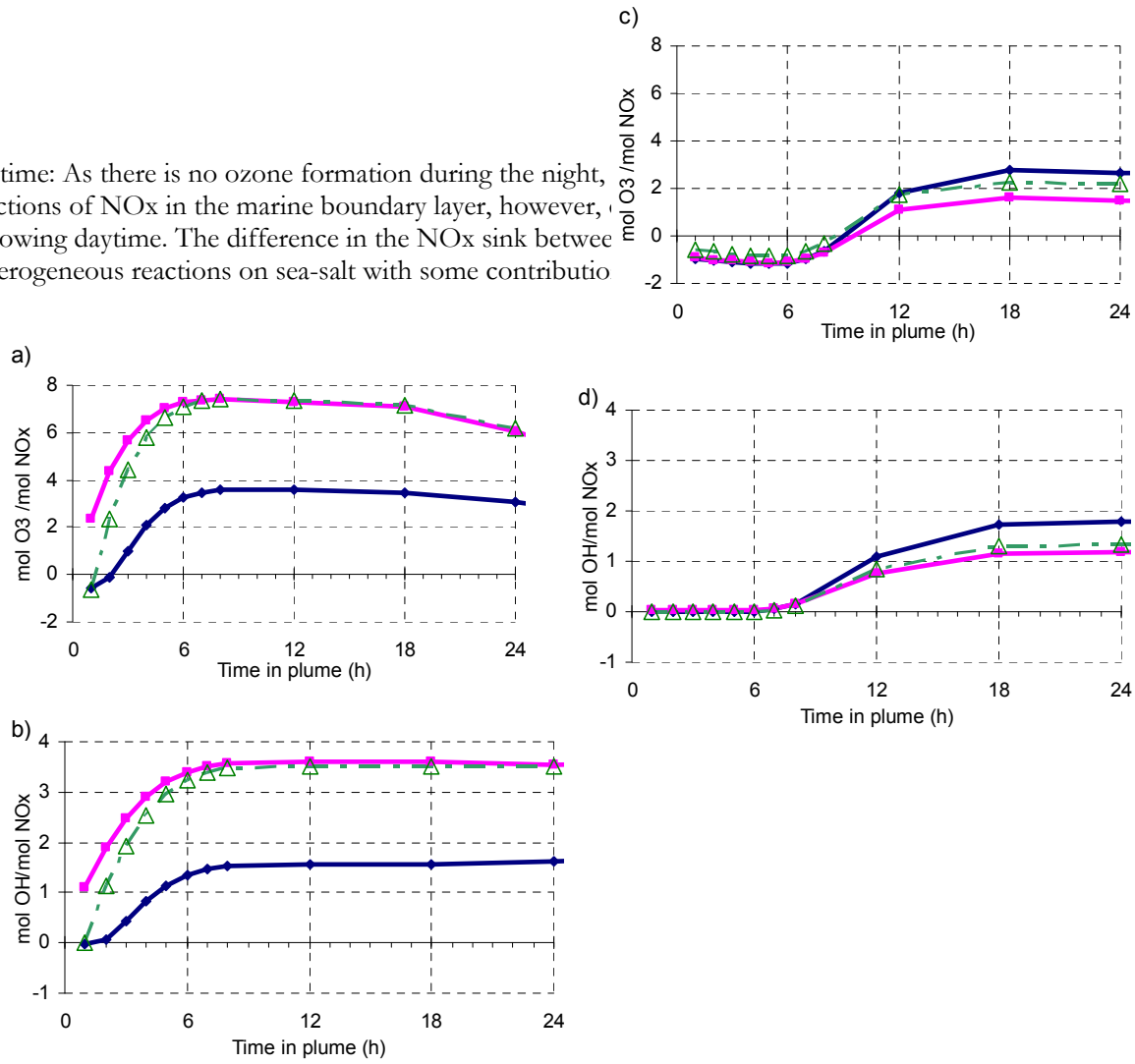


Figure 1. Molar mass of ozone formed in the plume released at daytime (a), OH formed in the plume released at daytime (b) ozone formed in the plume released at nighttime (c) and OH formed in the plume released at nighttime (d), relative to the molar mass of emitted NOx in the different model simulations. Dark blue line – ED simulation, purple line – ID simulation, dashed green line – IDP simulation parameterised with a nonreactive plume-NOx tracer, green triangle – IDP simulation parameterised with tracer depleting ozone by rate Keff.

The heterogeneous reactions on background sea-salt and sulphate particles are important sinks of NOx and reduce ozone formation both in the ship plume and in the background marine air. In a plume released after the dusk 50 – 60% of NOx is removed by heterogeneous reactions while in a plume released in daytime the heterogeneous reactions removed 18 – 20% of the emitted NOx (Figure 2). The nonlinearities in NOx sink are dominated by the NO2 + OH reaction at daytime and by heterogeneous reactions at night. Figure 3 shows difference in normalised sink of the emitted NOx between the ID and ED simulations.

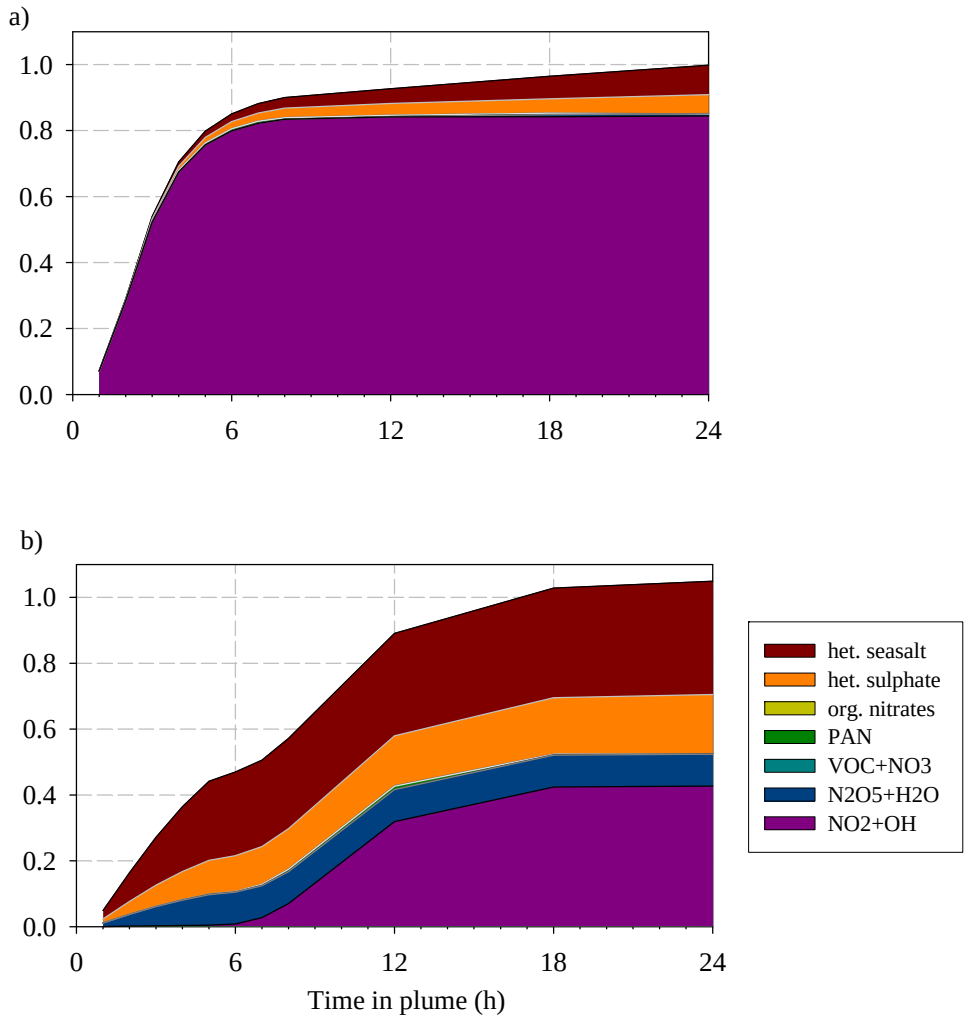


Figure 2. Molar mass of plume NOx, normalised by the NOx emission, oxidized by different reactions; a – in plume released at daytime; b – in plume released at night.

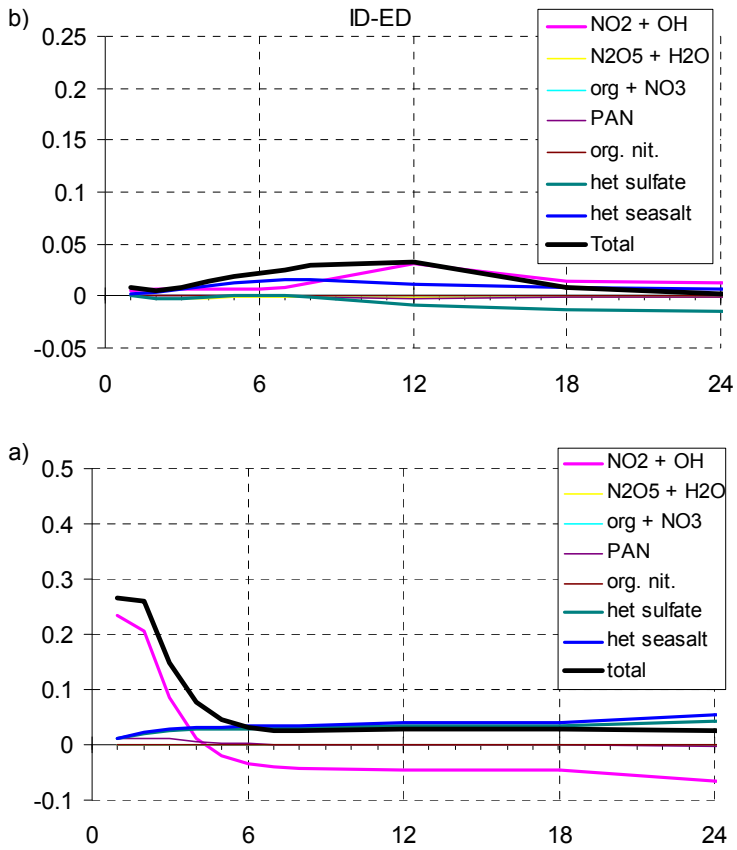


Figure 3. Difference between the normalised sinks of NOx in the ID and ED simulations; a – plume released at daytime, b – plume released at nighttime.

For SO_2, scavenging on sea-salt particles followed by aqueous-phase oxidation contributed by 6 – 8% to the total SO_2 sink (over 60% was dry deposition on the sea surface) (Figure 4).

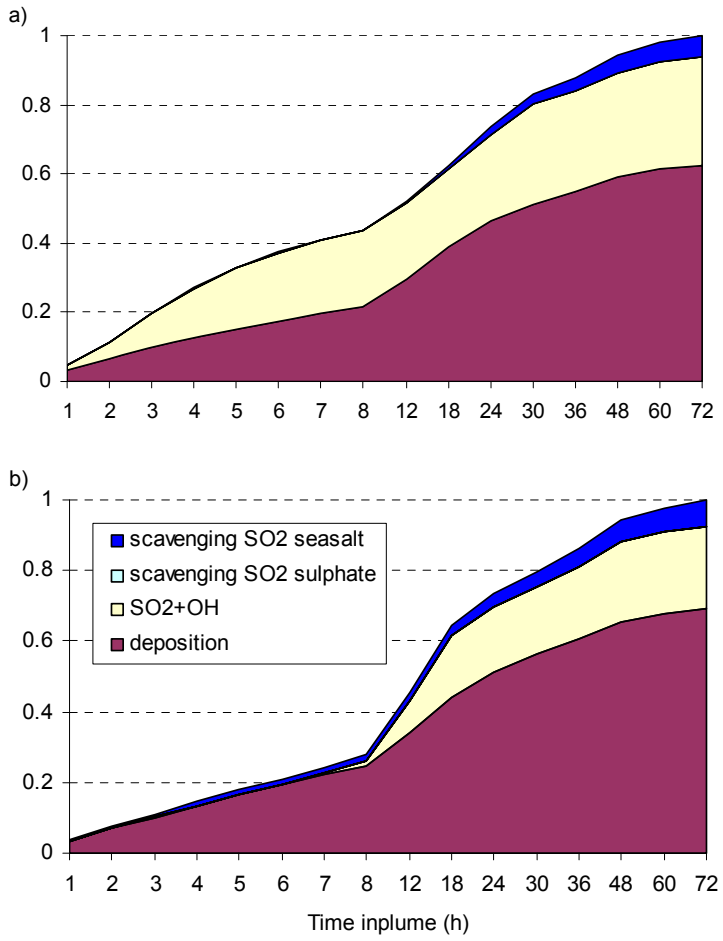


Figure 4. Difference between the normalised sinks of SO_2 in the ED simulations. a – plume released at daytime, b – plume released at nighttime.

As can be seen in Fig. 1, there is a need to reduce potential of NOx to form ozone. If the plume-NOx tracer reacts with OH (ozone must be titrated for that part of NOx that was emitted as NO) we obtain a direct sink of NO_2 and OH without ozone formed under the reaction cycle (simulation IDP Keff2). The lifetime of the plume-stage NOx becomes in this type of simulation a very important parameter for the performance of the parameterisation. Figure 5 shows budgets of reactions on accumulated ozone formation in plume (a) and accumulated NOx sink for several lifetimes of the plume-stage NOx tracer. We can see an improvement of performance of the parameterisation and rather good agreement in case with τ=140 min..

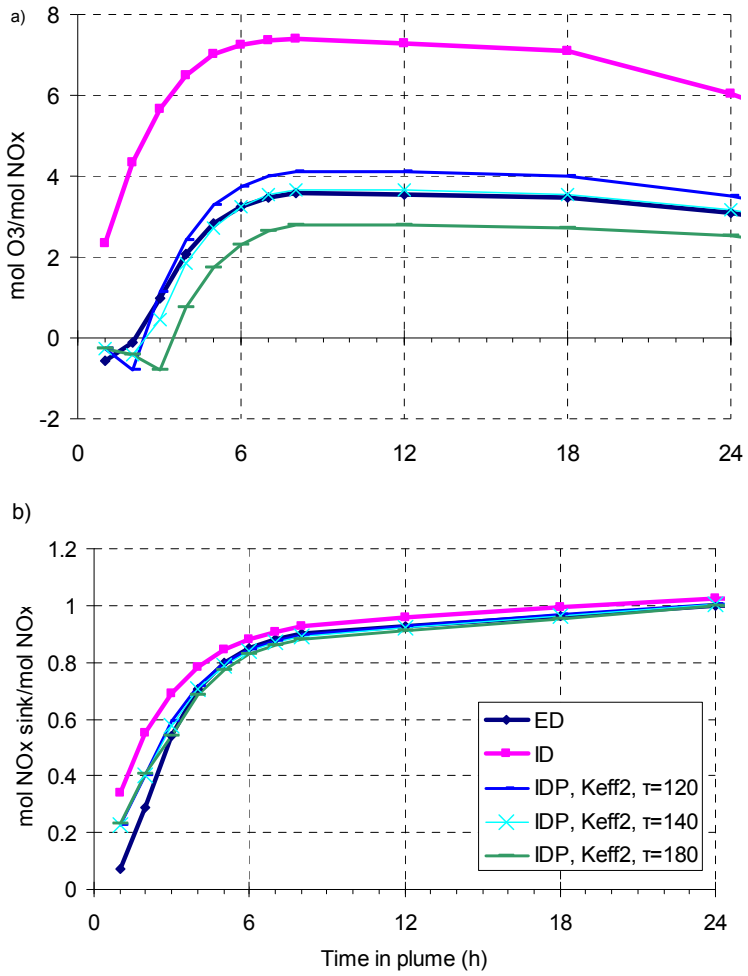


Figure 5. Accumulated budgets of ozone sources (a) and NOx sinks (b), normalised by molar mass of NOx emitted, simulated for the ID and ED cases and for 3 parameterised IDP cases: IDP, Keff2, τ =120, parameterisations with tracer reacting with OH and with τ = 120 min; IDP, Keff2, τ = 140 and τ = 180 same but the plume-stage NOx lifetime is 140 and 180 min, respectively.

The sensitivity of the performance of parameterisation to the part of the plume-stage NOx tracer removed (which is proportional to its lifetime τ) can be seen in Figure 6. We can see the different sensitivity of plumes released at dark.

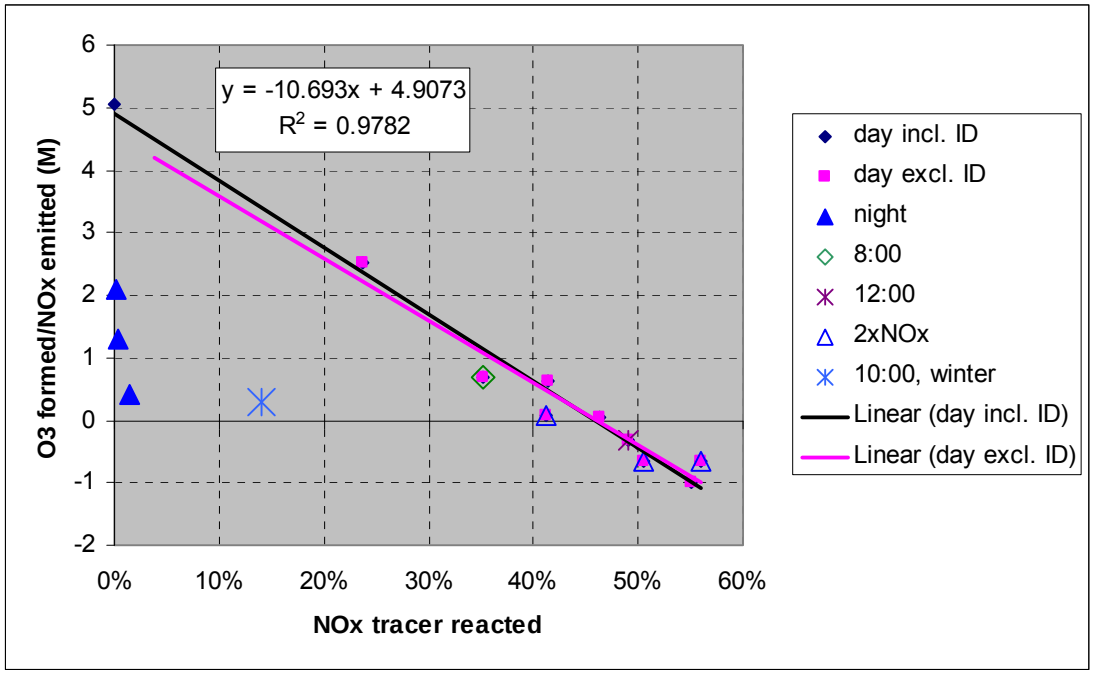


Figure 6. Difference of parameterised IDP Keff2 simulations from ED in normalised ozone formation, plotted against mass of NOx tracer that reacted with OH (rate Keff2): same symbols – test of different values of τ (in minutes)

4 Conclusions

- Parameterisation with sink for NOx tracer + OH behaves well in terms sensitivity of τ to tested conditions. At nighttime tracer sink need to be treated in a different way taking into account the heterogeneous reactions.
- There are 2 possibilities how to parameterise: use of Keff = k(OH+NO_2)*OH*k2, k2 comes from the mixing ratio of the tracer, or to calculate β as a function of OH.
- Statistics of the plume dispersion at MBL needs to be studied and the functional dependence between statistics of the mixing lifetimes and corresponding τ needs to be studied.

4.1 References

Cariolle, D. et al. (2009). Parameterization of plume chemistry into large-scale atmospheric models: Application to aircraft NOx emissions, J. Geophys. Res. 114, D19302, doi:10.1029/2009JD011873.