Periodic Activity report month 25-36

Version 1 April 2008

2.3 Activity 3: Large-scale Chemistry Effects (Isaksen, Van Velthoven)

2.3A Summary of progress

Objectives

- To estimate the current change in chemical composition from different modes of transport (O.3.1.2.2) and thereby to provide input data on change for impact studies (O.3.1.2.1).
- Similarly, to estimate the changes in future chemical composition (O3.3.1.2), and to estimate the effect of atmospheric chemistry-climate feedbacks on the future impact of emissions from transport (O3.3.2.1).
- To evaluate the latest versions of the models against observations focusing on key processes for correct simulation of the effects of transport emissions (O.3.1.1.1).
- To define or select suitable effective emissions for aviation, shipping and road transport (O3.1.3.1), to apply these emissions in the ACMs (O3.1.3.2), and to estimate the effect of sub-grid scale chemistry and dispersion on the chemical perturbations from transport (O3.1.3.3).
- To assess the contributions from transport modes to wet and dry deposition (O3.2.1.2).
- To estimate the effect of cirrus clouds on the impact of transport emissions (O3.2.3.1).
- To investigate options for reducing the impact of individual transport modes and regions (O3.3.3.1)

Progress towards objectives

Model evaluation has been carried out for the year 2003 comparing model results with observational data on a point-to-point basis using data from the SPURT campaign and the MOZAIC program. UT/LS ozone is generally very well correlated with the SPURT and MOZAIC data. Correlation coefficients for the SPURT campaigns range from 0.8-0.9 in July to ≥ 0.9 in February. The comparison with MOZAIC reveals that correlations are high in the extratropics in the winter season (≥ 0.8), somewhat lower in summer (≥ 0.75 over large parts of the northern hemisphere), and low the subtropics and tropics (< 0.6). Mean model biases are mostly moderate for the SPURT campaign (< ± 30 %). All models, but MESSy show positive ozone deviations from MOZAIC observations over the entire northern hemisphere in winter. In contrast, in summer, while subtropical and tropical UT/LS ozone is generally overestimated by all models, in the extratropics the models mostly show negative biases.

Concerning carbon monoxide (CO), all models, but OSLO CTM2 using POET emissions, significantly underestimate the values observed in SPURT in the middle and upper troposphere, while good agreement is found in the lowermost stratosphere (Fig. 2.3.1). In contrast, OSLO CTM2 (POET) agrees well in the upper troposphere, but overestimates CO in the lower stratosphere. On the hemispheric scale, comparing model output with MOZAIC data at 300-170 hPa, biases can be summarized as follows: TM4, pTOMCAT T42, and MESSy generally show moderate to large negative biases during all seasons of the year 2003, while MOCAGE and LMDzINCA exhibit spatially and seasonally varying biases. During summer, all models (except OSLO CTM2 (POET)) underestimate observed CO.

Modelled CO abundance largely depends on the specific emissions inventories used, as can be clearly seen from Fig. 2.3.2, which shows mean model biases of different simulations with OSLO CTM2. While UT/LS CO mixing ratios are significantly overestimated when using

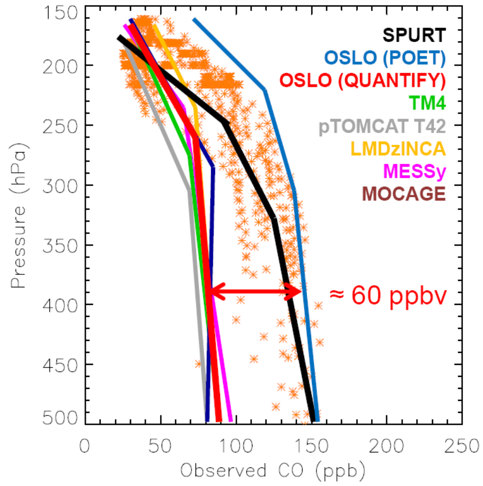

Fig. 2.3.1: The February 2003 SPURT campaign: Observed and modelled vertical CO profiles (ppbv) as a function of pressure (hPa). SPURT measurements (orange stars), estimated mean observational profile (thick black line). Estimated model profiles: UiO with POET emissions (blue), UiO with preliminary QUANTIFY emissions (red), TM4 (green), pTOMCAT T42 (grey), LMDzINCA (yellow), MESSy (pink), and MOCAGE (brown).

POET anthropogenic, biomass burning and road emissions, much smaller biases, that may even change sign over large regions (Fig. 2.3.2b) occur when using the EDGAR FT2000 inventory for anthropogenic emissions, GFED estimates for biomass burning, and road emissions provided by AC1. In fact, OSLO CTM2 then simulates CO abundance consistently with the other models also underestimating the observed mixing ratios at lower altitudes (Fig. 2.3.1).

Contributing factors to differences between OSLO CTM2 using POET emissions and the EDGAR/GFED/QUANTIFY emissions may be that POET anthropogenic NMVOC emissions are a factor 2 higher than other emission inventories (e.g., EDGARFT2000) over polluted regions such as Europe. In addition, tropical biomass burning emissions based on GFED are representative for the year 2000, and may thus not be characteristic for 2003. Some smaller dependency on the road emissions inventory used is also evident from Fig. 2.3.2b and c. All, but one model have used QUANTIFY preliminary road emissions that amount to a global mean of 73 Mt CO/yr. The updated road emissions that have been available lately, are increased and globally amount to 110 Mt CO/yr.

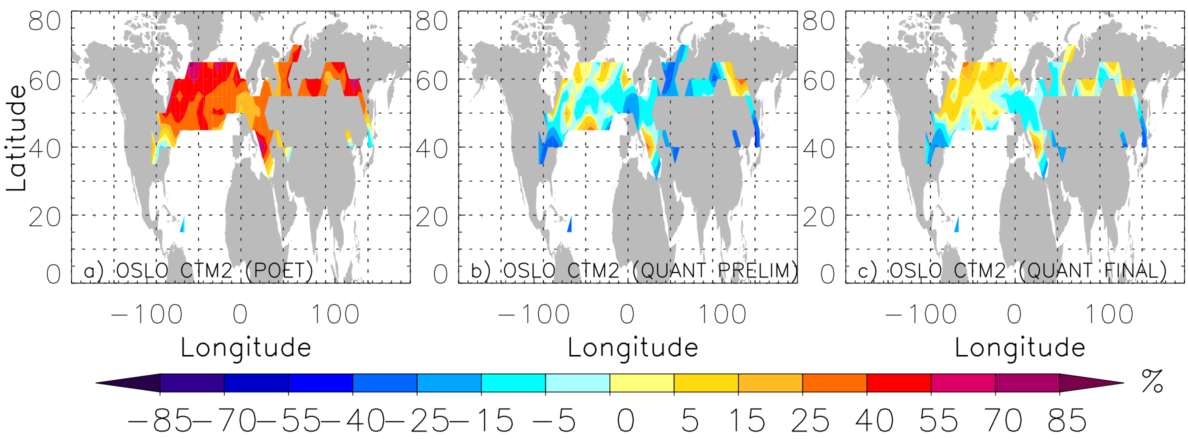

Fig 2.3.2: Mean model bias of OSLO CTM2 model compared to MOZAIC at 300-170 hPa for MAM 2003. a) using POET emissions, b) using QUANTIFY preliminary emissions, and c) using QUANTIFY final emissions.

The simulations of the current impact of the emissions from the transport sectors have been completed and the report summarising the impacts (D3.1.2.9) has been submitted. The mean maximum effect of emission reduction of traffic emissions predicted by the models is 4.0 DU and maximizes over the northern subtropical Atlantic (see Fig. 2.3.3). The relative effect amounts to more than 15% in the zonal mean (5.5 ppbv) compared to unperturbed conditions

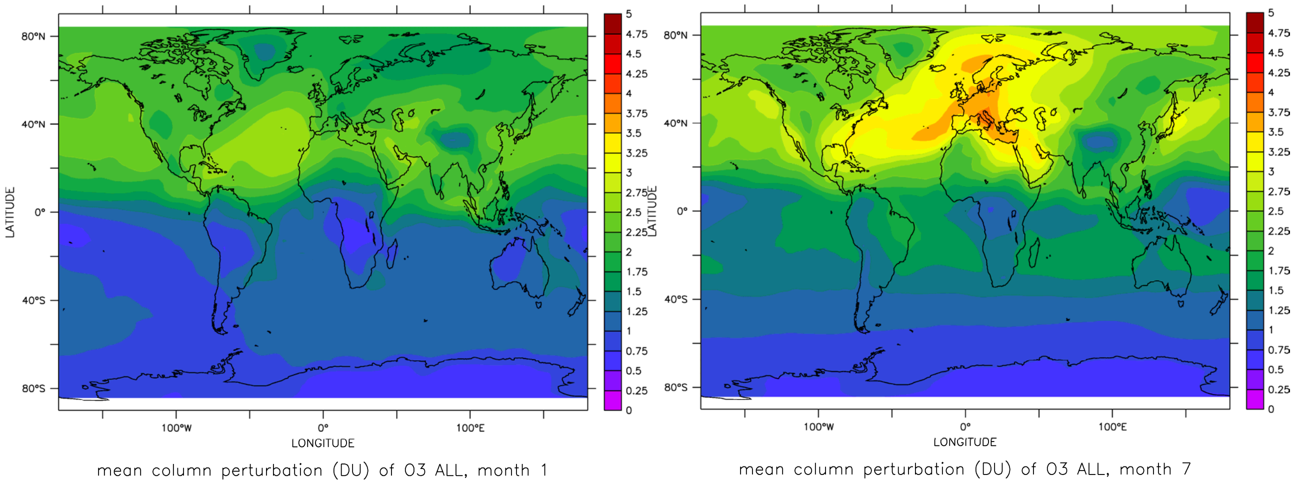

Fig.2.3.3: Mean column ozone perturbation (DU) due to the summed effect of road, ship and aircraft emissions for January (left) and July (right) (mean of the ensemble of models scaled to 100%)

In the region of maximum column ozone sensitivity, the emissions of ship contribute to more than 80% to the boundary layer during northern summer. Over the southern hemisphere troposphere ship emissions contribute to the total ozone perturbation from 60%–80% throughout the year (1–1.5 ppbv, Figure 2.3.4).

Road emissions have the strongest impact on ozone over the continental boundary layer and the free troposphere. Their impact on the northern hemisphere upper troposphere competes with aircraft emissions over the continents due to vertical transport and convection. Investigation of the efficiency of the NOx perturbation on ozone shows that aircraft emissions have the strongest impact on ozone in the tropopause region.

With regard to the OH budget, ship emissions perturb the zonal mean by 10–16% (zonal mean), which can exceed $5 \cdot 10^5$ molecules/cm^3. Methane lifetime reduction due to ship emissions is estimated to be on the order of 3.5–4%, for road traffic emissions about 1.5%, and for aviation emissions around 1%.

Output fields (O_3, CH_4, OC, BC, SO_4) from the simulations of current impact have been made available to Activity 6 for radiative forcing calculations.

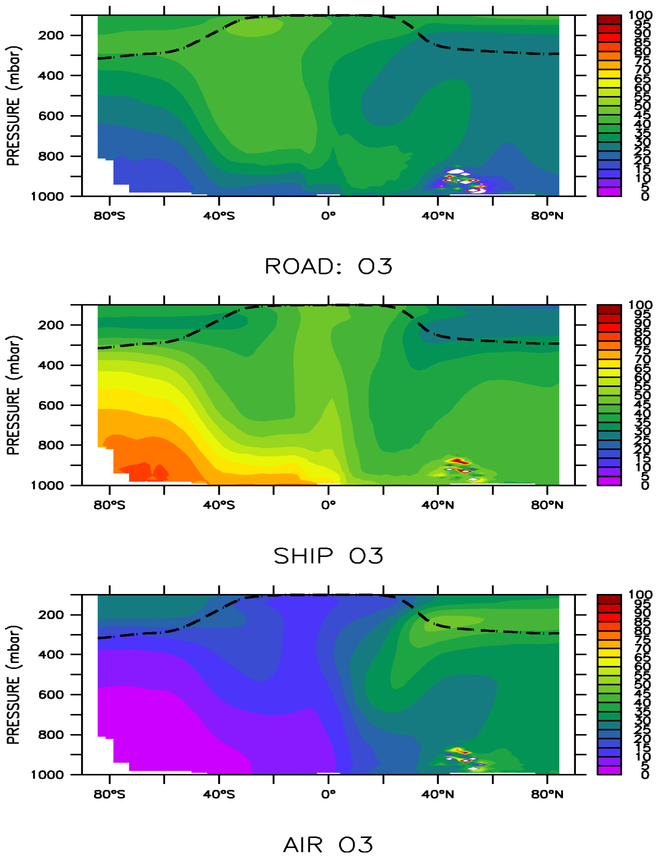

Fig.2.3.4: Relative fraction of each type of traffic to the total ozone perturbation for January (mean of the ensemble of models scaled to 100%)

Within QUANTIFY there has been quite some discussion about the meaning of 5% perturbations. For this reason, UCAM has performed supplementary simulations with 100 % perturbations of the transport sector emissions instead of the 5% perturbations of the original experimental set-up. While for emission perturbations of up to 20 % the ozone and methane perturbations vary linearly with the emission perturbation, this is not the case anymore for 100 % perturbations. This result is not unexpected: it was the reason we originally used 5% perturbations. We would like to stress that 5% and 100% perturbations essentially provide different information. 100 % perturbations give the impact that totally switching off a certain transport sector would have, while 5 % perturbations allow us to estimate the impact of a certain (less than about 20%) amount of emissions from aviation, road traffic or shipping under present day circumstance if all other emissions remain the same. When using 100% perturbations the sum of the individual impacts from aviation, road traffic and shipping is different from the impact of the sum of these emissions (non additiveness due to non-linearity). An impact thus only has meaning when one specifies the context of all other emissions.

Figure 2.3.5 shows for each of the sectors the percentage difference in ozone impact between 20 times 5 % perturbations and 100 % perturbations. The relative difference is largest for road traffic, which is relatively the largest source, and geographically in the southern hemisphere because of its relatively clean background. Table 2.3.1 shows the corresponding differences in ozone radiative forcing and methane lifetime. Especially, the estimate for the change in methane lifetime depends critically on how the perturbation is defined (what the background is).

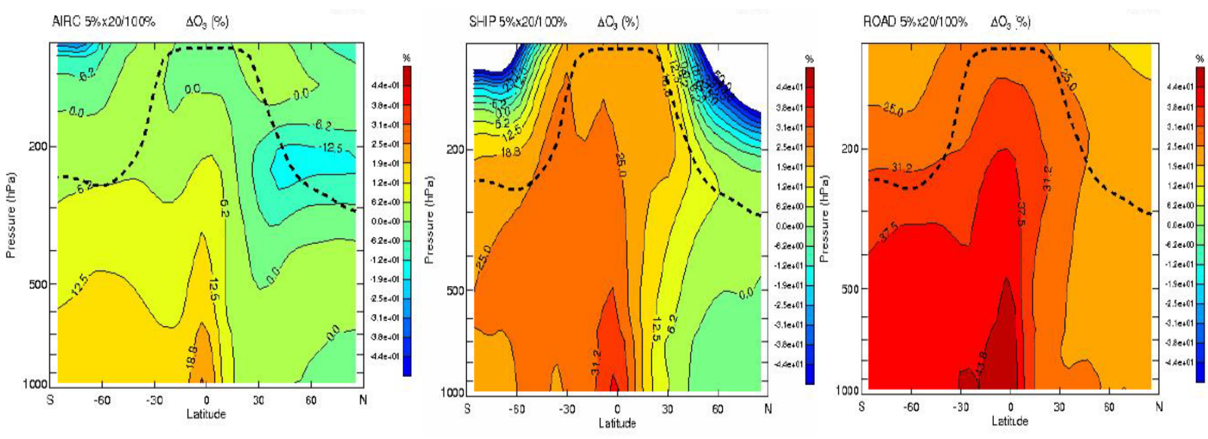

Fig.2.3.5: Percentage difference in the impact of aircraft, ship and road traffic emissions on ozone between 20 x 5% perturbations and 100% perturbations. The results are from p-TOMCAT.

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	Aviation	Shipping	Road traffic
Ozone RF 20x5%	28.0	23.8	15.9
Ozone RF 100%	24.8	18.0	11.0
Δτ (CH4) 20x5%	-0.24	-0.68	-0.20
Δτ (CH4) 100%	-0.13	-0.38	-0.11

Table 2.3.1: Comparison between 20 x 5% perturbations and 100% perturbations for ozone radiative forcing (mW/m2) and the change in methane lifetime (years). The results are from p-TOMCAT.

The uncertainties due to plume processing are being estimated by calculating the difference in pollutant concentrations between ACM simulations with and without effective emissions. During the third year of the project, CNRS-LSCE has implemented the new CERFACS approach (D3.1.3.1) for parameterisation of sub-grid scale dispersion and chemical conversions in the LMDz-INCA model (D3.1.3.2). A first report with estimates of the influence of sub-grid scale chemical conversions on the impact of aviation emissions (D3.1.3.8) has been provided. Results show that the introduction of the non-linearity in the atmospheric chemistry during the aircraft plume dispersion leads to NO_x and associated O_3 decreases along the major aircraft routes: Up to 5% (July) and 15% (January) of attenuation of the O_3 production from aircraft emissions is obtained in the high troposphere of Northern Hemisphere (Fig. 2.3.6).

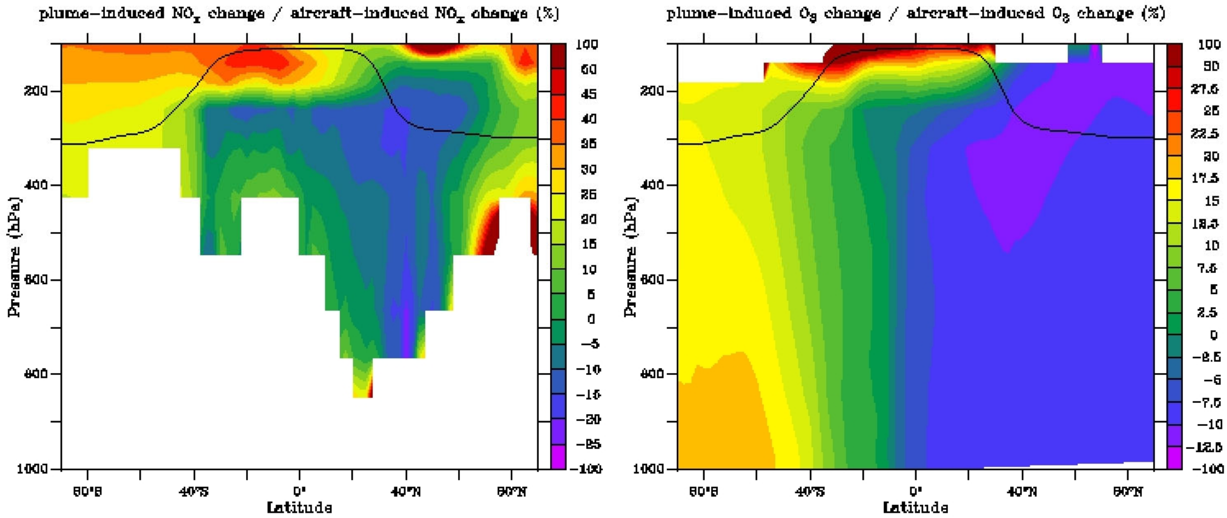

Figure 2.3.6: Zonally averaged relative changes (%) in the impact of aircraft emissions on NO_x (left) and O_3 (right) concentrations due to plume effects, for January and an assumed plume lifetime of 15h.

The model intercomparison of removal of nitrogen compounds has now been finished and the report (D3.2.1.5) has been submitted. This compares the magnitude and distribution of dry and wet removal of NO_y compounds resulting from NO_x emissions from the individual transport sectors (road traffic, shipping, aircraft). In total, 5 models have participated in the deposition intercomparison. Apart from the originally planned TM4 (KNMI), LMDZ-INCA (CNRS-LSCE) and p-TOMCAT (UCAM) models, also the E39C (DLR) and MOCAGE (CNRM) model provided output to the database for this experiment. In most models, removal of NO_y from all sources, including transport emissions and lightning, exhibits a slight preference for wet deposition. In contrast, NO_y compounds related to the transport NO_x emissions seem to be preferentially removed through dry deposition, presumably because these emissions are largely surface-based. However, uncertainties related to this subdivision, as deduced from intermodel differences, are substantial. As an illustration, figure 2.3.7 shows results from 3 models for deposition of NO_y originating from ship emissions.

Generally, NO_y constituents are preferentially deposited in the regional vicinity of the NO_x-sources, which in case of the transport-related emissions of NO_x means the (northern hemisphere) congested regions and the global transport routes. There is considerable uncertainty concerning the magnitude and location of the peak deposition rates for some transport modes. Based on intermodel differences in NO_y deposition of transport-induced emissions of NO_x we could conclude that the uncertainty in wet deposition distribution is higher than that in dry deposition, in particular with regard to the regional-scale features. This is because the relevant processes involved in deposition of NO_y (transport and removal in cloud systems) act mainly on regional scales, and the description of these processes differs considerably among the models. Furthermore, not unexpectedly, the agreement improves when going to (averaging over) larger spatial scales.

With transport-induced NO_x-emissions being projected to rise disproportionately compared to other sources in the coming decades, the model intercomparison has helped to identify regions that will potentially be most affected by the increasing surface deposition of nitrogen compounds, i.e. the coastal regions of western Europe and the U.S. and the east coast of Asia.

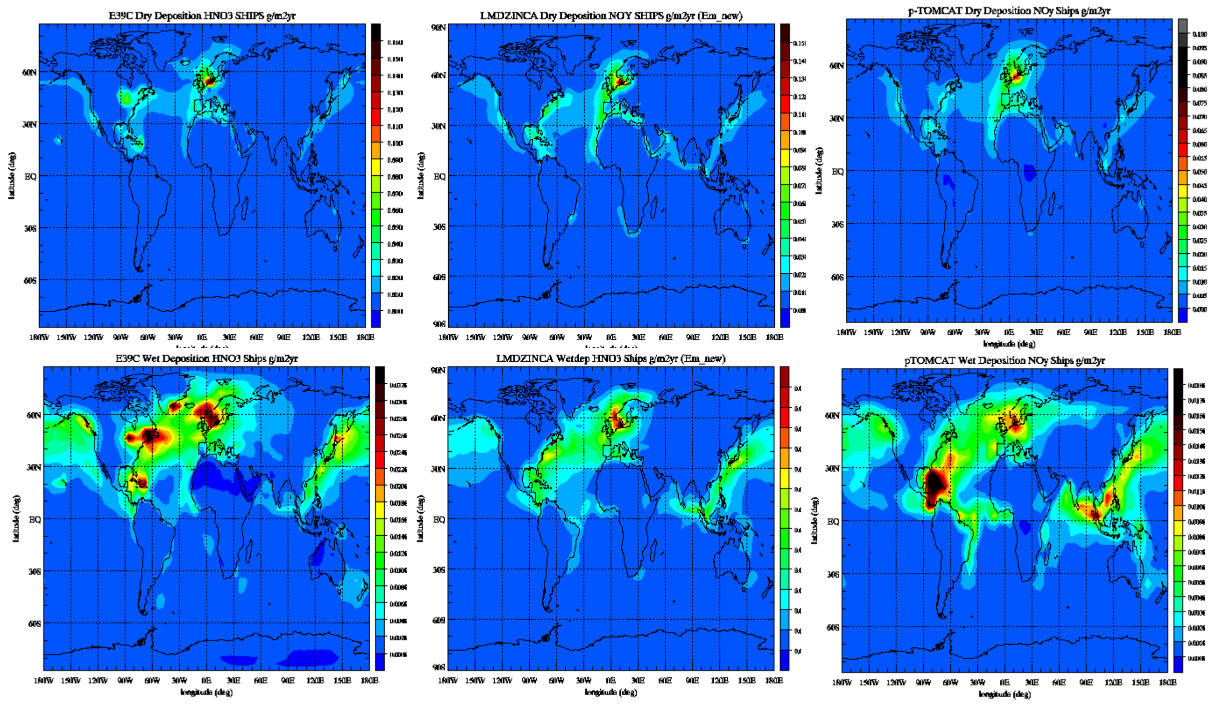

Figure 2.3.7: Dry (upper panels) and wet deposition (bottom panels) of NO_y originating from ship emissions as simulated by the E39C, LMDZINCA and p-TOMCAT models.

Deviations from the project work programme

WP3.1.1 Evaluation of multimodel performance

ETH has requested a shift of the final report on model evaluation to month 48 to enable them to spend all planned budget, since their postdoc has had unexpected periods of leave (related to pregnancy). The coordinator of QUANTIFY has already agreed to this shift.

WP3.1.2 Current impact

Due to remaining problems with the coupling between chemistry and dynamics in the MPICHEM model, deliverable D3.1.2.10 concerning calculated perturbation fields was found not be scientifically reasonable. Instead a detailed simulation in high resolution mode of the measurements performed in Activity 5 will be performed in year 4 by MPICHEM. It must be noted that MPICHEM, as leader of WP3.1.2, has spent quite a lot of effort on the analysis of the output from the whole set of models, and the reporting of this WP, including the preparation of a joint paper. Future work of MPICHEM will be redirected towards simulation of the ship measurement campaign. We also note that output from additional model (MOCAGE from CNRM/Meteo-France) has been used in the analysis. Hence the total number of models contributing model output is as planned.

WP3.1.3 Effective emissions

The proposal for a new approach for treating sub grid scale plume effects was delivered last year by Activity 2. Preliminary tests with this parameterisation in the LMDZ model, performed in month 24-30, yielded a very small impact of plume processing on the ozone perturbations caused by aviation. In month 30-36 the parameterization has been adapted to include some missing chemical reactions, so that presently results have been obtained that compare well with alternative approaches published in the literature. In our view it is quite satisfactory that conceptually quite different approaches yield similar results. In view of other delays, e.g. with WPs 3.3.1-3 we are planning not to give priority to implementation of this new plume parameterisation scheme in further models. In the end it is more interesting to have a comparison of fundamentally different approaches, than a comparison of results of implementation of the same parameterization in different models. The former gives a more comprehensive estimate of uncertainty. We will, however, still implement the new parametererization in the UIO model since this allows direct comparison of the new CERFACS approach to the older (Kraabøl) approach. This is scheduled for month 43-48 since the dependency of Act. 6 on output from WP3.3.1-2 requires us to give priority to those WPs (see below).

WP 3.2.1 has been finished.

WP 3.2.2 is ahead of schedule, since UCAM already finished a large part of the simulations due month 44.

WP 3.2.3 Impact of cirrus clouds

As reported last year KNMI suffered from a delay in WP3.2.3.and WP3.2.1 due to the premature departure of their postdoc. In the meantime a new postdoc has started at KNMI and WP3.2.1 has been finished. However, most of the work in WP3.2.3 still has to be done. This will be scheduled in the second half of the 4th year, since priority needs to be given to the simulations for WP3.3.1 on future composition changes because Act. 7 depends on this output (see below). A prolongation until e.g. month 54 of QUANTIFY might be useful to fully exploit the work in this WP.

WP3.3.1-3 Future composition changes, impact of climate change and evaluation of selected mitigation options

These WPs have suffered delays due to the late delivery of gridded emission scenarios by Act. 1. Most of the gridded data were delivered in month 33, and in month 35 in the case of aviation. The aviation files are still being updated after it was decided on the third annual meeting in Budapest in February 2008 that the scaling should be included in the gridded files. These gridded files still needed and need further preprocessing (regridding, grouping of constituents and re-scaling) and testing by the modelling groups which is currently taking place. It is now planned that the runs for WP3.3.1 and WP3.3.2 take place until June 2008 (month 40) followed by analysis until month 42. The runs and analysis for WP3.3.3 will be done subsequently month 42-48. It is important for the RF and scenario calculations in Act. 7 that future perturbation fields from WP3.3.1 become available well before the end of the project.

2.3B Active work package descriptions

WP 3.1.1 Evaluation of multi-model performance against observations (Schnadt, ETH Zürich)

WP number	3.1.1	Period month 25-42		Start date :	1	End date	48
Participant id	DLR-IPA	ETH Zürich	CEA	KNMI	MPICHEM	UIO	
Person-months*	0 (0)	11.1 (6.9)	0 (0)	0 (0)	0 (0)	0 (1)	
Participant id	UCAM-DCHEM						
Person-months	0 (0)						

Objective

O.3.1.1.1 To evaluate the latest versions of the models against observations focusing on key processes for correct simulation of the effects of transport emissions (M3.1.1.3).

Description of work

Model simulations for 2003 have been defined and carried out including diagnostics of key processes relevant for the effects of the different modes of transportation. The model output is compared with observations from the established *ETH*meg observational database in order to evaluate the current performance of the ACMs.

The *ETH*meg Activity 3 model evaluation webpage (http://www.megdb.ethz.ch/index.php?site=

model_validation) will be updated regularly to include the most recent results from model evaluation with observations and model-model intercomparisons.

Model evaluation will be continued using the newest model versions. Focus of model evaluation will be stratosphere-troposphere exchange of ozone and NOx, boundary layer venting, and convective transport of pollutants from the surface. In case of DLR-IPA, for which only climatological model output can be delivered, a model evaluation is planned to be carried out using MOZAIC data from 2001-2004 to compare them against DLR model results of a transient integration of the years 1995-1999.

Description of progress

Point-to-point comparison of observational and model data has been carried out using the SPURT and MOZAIC observational data by ETH. Up to now, model temperature and ozone profiles have been compared to respective ozonesonde observations. The SPURT data have been used to evaluate model mean biases, tracer-tracer correlations, and taylor diagrams for the February, April, and July months 2003. Using MOZAIC data, horizontal distributions of UT/LS model biases and correlations have been computed. First results have been presented at international conferences and are used in a publication that is planned to be submitted (Hoor et al., 2008). UCAM-DCHEM has provided higher resolution model data (T42); UiO has delivered two reruns of the 2003 simulation using different emissions data sets. Evaluating these simulations give valuable indications of the effects of the use of different transport emissions, e.g. on the CO concentration. CNRM has provided diagnostic output for the MOCAGE model. Output diagnostics from DLR-IPA (D3.1.1.3) are still pending due to unexpected efforts needed for tuning of their new model.

The comparison of the model output with further observations (CONTRACE, surface data) is in progress. It is planned to evaluate the models with respect to near-surface observations from the QUANTIFY ship campaigns 2007 in autumn/winter 2008 when model simulations will have been repeated using the final QUANTIFY emissions data sets. Results can be viewed by the QUANTIFY partners at www.megdb.ethz.ch.

* man month numbers are given as: planned for month 25-42 (spent in month 25-36)

Deliverables

D3.1.1.9	First report on the performance of the models (month 25, ETH Zürich, delivered).
D3.1.1.10	Second report on the performance of the models (month 36, ETH Zürich, now planned for month 48)

Milestones

None

WP 3.1.2 Current chemical composition changes from different modes of transport (Lelieveld, MPICHEM)

WP number	3.1.2	Period month 25-42	Start date :	3	End date	48
Participant id	DLR-IPA	CNRS-LSCE	KNMI	MPICHEM	UIO	UCI
Person-months[*]	0 (1)	0 (0)	0(1)	3(2)	1 (3)	0 (3)
Participant id	UCAM-DCHEM					
Person-months	1 (2)					

Objectives

O.3.1.2.1 To provide input data on chemical change for impact studies

O.3.1.2.2 To estimate the current change in chemical composition from different modes of transport

Description of work

The report on the current perturbations, of which a draft version is available within Activity 3 will be finished. The MPI-GCM will offer a different analysis, since it was decided that a repetition of the previous runs in CTM-mode will not add significant progress to the work package. Instead MPICHEM offered to perform simulations of SO2 for current conditions (as well as for the future scenarios using IPCC sea surface temperatures for the future runs). It was discussed within Activity 3 and is believed to be the scientifically most satisfying solution rather than repeating the simulations which have been performed already by five other models.

The simulations of current impact were done with preliminary gridded emission inventories for transport that however hardly differ from the finally delivered ones. Yet, UCAM-DCHEM and UIO will perform additional simulations to quantify the consequence of these minor differences for the final large scale chemical perturbations. UCAM will do this only for the updated aviation emissions. UCAM will also perform an additional simulation with a 100% perturbation of road traffic emissions instead of the 5 % perturbation used in the original set-up, in order to get an upper estimate of the effect of non-linearity in the chemistry.

Description of progress

All participants of the workpackage have finished their runs and provided monthly mean output fields for 2003 to the Activity 3 data base webpage at the University in Oslo (http://norgrid.uio.no/mySRB). In addition to the original plan the data of the MOCAGE model of CNRM as well as from UCI are available now. The report (D.3.1.2.9) on the impact of traffic emissions on the current state of the atmosphere, which was delayed, has been made available by MPICHEM. It is the basis of a publication which is close to submission. It could be shown that the approach of a small perturbation by 5% to account for linearity of the effects on ozone was successful. Based on the small perturbation from the different types of traffic their relative contributions to the ozone perturbations have been evaluated. In addition the effect of traffic emissions for the OH budget in particular in the boundary layer could be shown. For the publication data analysis is continued including the results of the two additional models. Another paper focussing on results from the LMDZ model has also been drafted.

One group (MPICHEM) started to work on a high resolution model version to compare more detailed the measurements performed within Activity 5 with the simulations. Here, the focus is on a corridor effect rather than on single plumes. This approach replaces the simulations of SO2 for the current conditions since it turned out that the perturbation signal due to SO2 for the current conditions was too weak. It must be noted that MPICHEM has spent a lot of effort on the analysis of the output from the whole set of models, and the reporting of this WP, including the preparation of the joint paper.

UCAM has performed supplementary simulations with 100 % perturbations of the transport sector emissions instead of the 5% perturbations of the original experimental set-up. While for emission

[*] man month numbers are given as: planned for month 25-42 (spent in month 25-36)

perturbations of up to 20 % the ozone and methane perturbations vary linearly with the emission perturbation, this is not the case anymore for 100 % perturbations. This result is not unexpected: it proves that we were right in originally choosing 5% perturbations. It must be stressed that the 5% and 100% perturbations essentially provide different information. 100 % perturbations give the impact that totally switching off a certain transport sector would have, while 5 % perturbations allow us to estimate the impact of a certain (less than about 20%) amount of emissions from aviation, road traffic or shipping under present day circumstances. The impact on methane seems to be more sensitive to the precise definition of the perturbations than the impact on ozone.

Output fields (O_3, CH_4, OC, BC, SO_4) from the simulations of current impact have been made available to Activity 6 for radiative forcing calculations. The

Deliverables

D3.1.2.7	Preliminary current chemical perturbation fields from the UCI model (month 31, UCI, delivered month 34)
D3.1.2.9	Report with first-order estimate of the current impact on atmospheric composition (month 25, MPICHEM, delivered month 25)
D3.1.2.10	Chemical perturbation fields from the MPICHEM model in GCM-mode including SO2 (month 44, MPICHEM – will be modified in next DOW)
D3.1.2.11	Chemical perturbation field from p-TOMCAT for the final aviation emission inventory (month 36, UCAM-DCHEM – will be shifted month 42)
D3.1.2.12	Chemical perturbation fields from Oslo CTM2 for the final emission inventories (month 36, UIO, will be shifted month 42)
D3.1.2.13	Report with final estimate of the current impact on atmospheric composition (month 48, MPICHEM)

Milestones

None

WP 3.1.3 Implementation of effective emissions (Hauglustaine, CNRS-LSCE)

WP number	3.1.3	Period month 25-42	Start date :	19	End date	36
Participant id	CNRS-LSCE	CEA-LSCE	KNMI	MPICHEM	UIO	UCI
Person-months*	12 (10)	0 (0)	5 (0.2)	4(0)	3 (1)	4 (0)
Participant id	UCAM-DCHEM					
Person-months	4 (0)					

Objectives

O3.1.3.1	To define or select suitable effective emissions for aviation, shipping and road transport
O3.1.3.2	To implement apply these emissions in the ACMs
O3.1.3.3	To estimate the effect of sub-grid scale chemistry and dispersion on chemical perturbations from aviation, shipping and road transport

Description of work

Currently emissions by transport activities are usually directly emitted into the coarse grid cells of global ACMs. These grid cells have sizes of typically 100-500 km. The uncertainty due to plume processing will be estimated by calculating the difference in pollutant concentrations between ACM simulations with and without effective emissions. For aviation the large uncertainty range in published calculations of effective emissions needs to be reduced. Preliminary estimates of aircraft plume processing have been made by UIO and KNMI. New effective emissions for shipping and aviation will be based on a parameterisation developed by CERFACS in Activity 2 that makes use of additional large scale fuel tracers for aviation and shipping. Coefficients for this were supplied by CERFACS in month 23. CNRS-LSCE is implementing and evaluating this new approach in their ACM. This will take about 3 months. The new effective emissions will them be implemented in several ACMs because differences due to model formulation need to be taken into account in the estimate.

Description of progress

The uncertainties due to plume processing have been estimated by calculating the difference in pollutant concentrations between ACM simulations with and without effective emissions. During this third year of the project, the CNRS/CEA-LSCE has implemented the CERFACS new approach (D3.1.3.1) in the LMDz-INCA model (D3.1.3.2) and has provided a first report on estimates of chemical perturbations from aviation sub-grid scale chemical conversions (D3.1.3.8). Several simulations have been carried out to test performances of the new plume parameterization considering a fuel tracer. A run introducing the plume parameterization with a plume lifetime of 1s and without the small-scale ozone loss terms in the large scale prognostic equation for ozone, has been performed to determine the capacity of the new parameterization to produce results consistent with those given by the base case run when the plume lifetime is reduced to small values. Another run was carried out with the required plume lifetime of 2h but still without the small-scale ozone loss terms. The last run was finally the one prescribed in the description of the CERFACS approach in the deliverable 3.1.3.1. The plume lifetime has been set to 15 hours, so that it includes both the initial phase, where the non-linearity of the tropospheric ozone chemistry is high, and a latter one related to the duration of the plume dispersion. A K_{eff} coefficient of 3.10^{-18} molecules^{-1} s^{-1} cm^{3} has been used, as deduced by using a box model exploring a large range for NO_x loading. Results show that the introduction of the non-linearity of the atmospheric chemistry during the aircraft plume dispersion leads to NO_x and associated O_3 decreases along the major aircraft routes: Up to 5% (July) and 15% (January) of attenuation of the O3 production from aircraft emissions is obtained in the high troposphere of Northern Hemisphere. The method will still be implemented in the Oslo CTM2 model by UIO during the second half of 2008 in order to take into account the differences due to the model formulation. Because of delays in the development of the parameterization, only the two above mentioned models will provide the related

* man month numbers are given as: planned for month 25-42 (spent in month 25-36)

output fields. Hence deliverables D3.1.3.3, D3.1.3.4, D3.1.3.5, D3.1.3.7 will be cancelled, and D3.1.3.6 will be shifted to month 48. For the same reason, only the perturbations from aviation plumes will be studied and reported. The results are however being compared to published results from alternative plume parameterisations made by KNMI and UIO.

Deliverables

D3.1.3.2	Output fields from ACM simulations with/without effective emissions (month 34, CNRS-LSCE, delivered month 36).
D3.1.3.3	Output fields from ACM simulations with/without effective emissions (month 34, KNMI, cancelled).
D3.1.3.4	Output fields from ACM simulations with/without effective emissions (month 34, MPICHEM, cancelled).
D3.1.3.5	Output fields from ACM simulations with/without effective emissions (month 34, UCAM-DCHEM, cancelled).
D3.1.3.6	Output fields from ACM simulations with/without effective emissions (month 34, UIO, will be shifted to month 48).
D3.1.3.7	Output fields from ACM simulations with/without effective emissions (month 34, UCI, cancelled).
D3.1.3.8	Report on the uncertainty in estimates of chemical perturbations from aviation and shipping due to sub-grid scale chemical conversions (month 36, CNRS-LSCE, delivered month 37)

Milestones

M3.1.3.1	Selection and setup of effective emission data sets / parameterisations performed (month 27, CNRS-LSCE, done)
M3.1.3.2	Model simulations with/without effective emissions carried out (month 34, CNRS-LSCE, done month 34)

WP 3.2.1 Precipitation impact on sulphur and nitrogen compounds (Van Velthoven, KNMI)

WP number	3.2.1	Period month 25-42		Start date :	8	End date	36
Participant id	CNRS-LSCE	KNMI	UCAM-DCHEM				
Person-months*	0 (0)	1 (3.5)	0 (1)				

Objectives

O3.2.1.1 To compare different parameterisations of removal of sulphur and nitrogen pollutants by precipitation (M3.2.1.2)

O3.2.1.2 To assess the impact of removal of sulphur and nitrogen compounds by precipitation on the impact of emissions from transport modes (M3.2.1.3)

Description of work

The parameterisation of wet deposition in the KNMI TM model has been evaluated and improved.

The sensitivity studies with the TM model indicated that the original set-up described in the first DoW of the project would not enable interpretation of the differences between the models. Hence, as described in the DoW for month 13-30, a simplified approach was followed. The 3 involved models have made simulations for 2003, where the wet and dry depositions were calculated for each of the 3 transport sectors separately. The report on these simulations (D3.2.1.5) still needs to be finished. The report is delayed because the postdoc employed by KNMI has left for another job.

Description of progress

The model intercomparison of removal of nitrogen compounds has now been finished and the report (D3.2.1.5) has been submitted. The report compares the magnitude and distribution of dry and wet removal of NO_y compounds resulting from NO_x emissions from the individual transport sectors (road traffic, shipping, aircraft). In total, 5 models have participated in this intercomparison. Apart from the originally planned TM4 (KNMI), LMDZ-INCA (CNRS-LSCE) and p-TOMCAT (UCAM) models, also the E39C (DLR) and MOCAGE (CNRM) model provided output to the database for this experiment.

Deliverables

D3.2.1.5 Report on the impact of removal of sulphur and nitrogen compounds on the chemical perturbations caused by transport modes (planned month 28, KNMI, submitted month 34)

Milestones

None

* man month numbers are given as: planned for month 25-42 (spent in month 25-36)

WP 3.2.2 Aircraft induced sulphur impact on UTLS ozone (Rogers, UCAM-DCHEM)

WP number	3.2.2	Period month 25-42	Start date :	33	End date	48
Participant id	CEA-LSCE	MPICHEM	UIO	UCAM-DCHEM		
Person-months[*]	2 (0)	2 (0)	2 (6)	6 (3)		

Objective

O3.2.2.1 To estimate the effect of aircraft sulphur emissions on UTLS ozone

Description of work

The impact on ozone of the increase in stratospheric aerosol density (SAD) due to aircraft sulphur emissions will be calculated. Recently published calculations of results from one model indicate (Pitari et al., 2002) that the downward transport of air depleted in ozone by the increased SAD is of the same order of magnitude as the ozone production from aircraft NOx emissions. This effect needs to be investigated with other models, because it is highly sensitive to vertical transport in the UTLS and to the microphysics implementation. A sensitivity study will be performed with several ACMs with perturbed and unperturbed stratospheric background aerosol climatologies.

At least one of the models will also compare a simulation of the perturbation caused by aircraft with background aerosol at post-Pinatubo levels with a simulation with the aerosol loading caused by Pinatubo.

Description of progress

The set-up of the model simulations has been fixed and described in a report (D3.2.2.1). UCAM has coupled the global stratospheric chemistry model SLIMCAT with an aerosol microphysical module (SAMM; Tripathi, 2004). This module simulates homogeneous binary nucleation, condensational growth, coagulation, and sedimentation of sulphuric acid-water particles with sizes from 0.3 nm to 1.5 mm. With this model UCAM has performed a 3 year run with NO_x, SO_2 and water vapour emissions based on AERO2K. The simulation period includes the Pina Tubo volcanic eruption causing a strong increase in background stratospheric aerosol density. The difference in the aircraft impact due to the increase in the sulphuric aerosol density has been calculated. UIO has also already performed preliminary simulations.

A cooperation with G. Pitari of the Un. Of Aquila has been set up on this subject. This had not been planned originally, but the coordinator of QUANTIFY has agreed to this cooperation and the corresponding extension of UIO man months in this WP.

In the coming months the simulations will be extended to more years and the remaining models will be involved in this WP. MPI-CHEM will not perform these simulations because of coupling problems.

Deliverables

D3.2.2.1 Report on the set-up of the model calculations of sulphur impact (month 35, UCAM, delivered month 30)

D3.2.2.2 Output from CEA model simulation with perturbed/unperturbed stratospheric aerosol (month 44, CEA)

D3.2.2.3 Output from MPICHEM model simulation with perturbed/unperturbed stratospheric aerosol (month 44, MPICHEM, will be cancelled)

D3.2.2.4 Output from UCAM-DCHEM model simulation with perturbed/unperturbed stratospheric aerosol (month 44, UCAM-DCHEM)

D3.2.2.5 Output from UIO model simulation with perturbed/unperturbed stratospheric aerosol (month 44, UIO)

[*] man month numbers are given as: planned for month 25-42 (spent in month 25-36)

D3.2.2.6	Report on the effect of aircraft sulphur emissions (month 48, UCAM-DCHEM)

Milestones

M3.2.2.1	Description available of model simulations to be performed (month 38, UCAM-DCHEM)

WP 3.2.3 Impact of cirrus clouds (Van Velthoven, KNMI)

WP number	3.2.3	Period month 25-42		Start date :	19	End date	36
Participant id	KNMI						
Person-months*	6 (2)						

Objective

O3.2.3.1 To estimate the effect of cirrus clouds on the impact of transport emissions

Description of work

Cirrus clouds are not only important for climate, but also for the UTLS ozone budget. Recent modelling studies suggest that chemical ozone loss on these ice particle surfaces is comparable in magnitude, but of opposite sign to aviation induced ozone production. Model studies have also demonstrated the significance of cirrus cloud particles for irreversible HNO_3 removal in the upper troposphere, significantly reducing NOx-catalysed chemical ozone production. However, only a few modelling studies on these issues have been performed so far. Recent laboratory research and observations, both in-situ and space-borne, have supplied new findings of key parameters that allow very useful following-up modelling efforts that have currently not been performed. These will be used to calculate in one of the ACMs the influence of cirrus on the impact of aircraft and other transport emissions on ozone. Where possible the numerical results will be evaluated with data from process studies resulting from other projects.

Description of progress

The new JPL (2006) recommendations for reaction rates have been implemented by KNMI in the TM4 model , leading to significant changes in some reaction rates (J_{MGLY}, $k_{OH\text{-}CO}$, J_{HCHO}, etc.).

Experiments with a modified ice cloud scheme are set up, including an evaluation of the parameterization of Kärchner&Lohmann (2002).

As reported last year KNMI suffered from a delay due to the premature departure of their postdoc. In the meantime a new postdoc has been hired. However, most of the work in WP3.2.3 still has to be done. This will be scheduled in the second half of the 4th year, since priority needs to be given to the simulations for WP3.3.1 on future composition changes because Act. 7 depends on this output. A prolongation until e.g. month 54 of QUANTIFY might be useful to perform the work in full depth.

Deliverables

D3.2.3.1 Description of a new parameterisation for ice cloud interaction based on recently determined parameters (month 30, shifted to month 38)

D3.2.3.2 Report on the effect of cirrus clouds on the chemical perturbations by transport (month 36, will be shifted to month 48)

Milestones

M3.2.3.1 A new parameterisation for ice cloud interaction based on recently determined parameters available (month 30, shifted to month 38)

M3.2.3.2 Model simulations carried out (month 34, will be shifted to month 46)

* man month numbers are given as: planned for month 25-42 (spent in month 25-36)

WP 3.3.1 Future chemical composition changes from different modes of transport (Isaksen, UIO)

WP number	3.3.1	Period month 25-42	Start date :	8	End date	48
Participant id	DLR-IPA	CNRS-LSCE	KNMI	MPICHEM	UCI	UIO
Person-months*	7 (1)	8 (2)	4(0)	3(1)	4(0)	8 (1)
Participant id	UCAM-DCHEM					
Person-months	3 (0)					

Objectives

O3.3.1.1 To provide input data of future chemical changes for impact studies (M3.3.1.4)

O3.3.1.2 To estimate the changes in future chemical composition from different modes of transport (M3.3.1.5)

Description of work

The global scale CTMs will be used to perform first-order studies of future chemical impacts. Changes in distributions will be calculated for O3, CH4 (through changes in its lifetime) and for particle distributions (sulphate, organic carbon aerosols). Time slice calculations for the target year 2050 will be performed with meteorology from 2003. These simulations will use gridded transport emission inventories for 2050 that will be delivered in month 30 by Activity 1. All modelling groups participating in the activities described in WPs 3.1.1 and 3.1.2 will participate in the activities. MPI-CHEM will perform these simulations in combination with current impact simulations described in WP3.1.2. DLR-IPA will only calculate perturbations for one specific transport sector (e.g. road traffic) and combine the results with simulations with the changed climate of 2050, as described in WP3.3.2.

The perturbed fields will be provided to WPs 6 and 7 for impact studies.

Compared to the original DoW WP3.3.3 has been slightly shifted forward in time, while WP3.3.1 has been shifted backward in time – in order to allow Activity 1 to provide emission input data in time.

Description of progress

Deliverables D3.3.1.2, D3.3.1.3, D3.3.1.3, D3.3.1.5, and D3.3.1.6 (future chemical perturbation fields from the CNRS-LSCE, KNMI, UCI, UCAM-DCHEM, and UIO models, respectively) were all due by month 36, but are delayed and will be delivered by month 40 (for one scenario at least). The reason for the delay is primarily the late delivery of emission scenarios, most of which were delivered in month 33, and in month 35 in the case of aviation. The aviation files are still being updated after it was decided on the third annual meeting in Budapest in February 2008 that the scaling should be included in the gridded files. ‘Scaling’ in this respect means an adjustment from pure ‘great circle’ routes to a scenario that assumes less efficient routing, said to be more realistic and featuring slightly higher emission totals.

All emission files were provided by Activity 1 in ASCII format. MPI has converted road and shipping emissions into netCDF format, which is used by Activity 3. The conversion includes consistent branching of VOC into the various hydrocarbon species that are used by the modellers. Aviation emission files were converted by UCAM. Non-transport emission files will be converted shortly.

For all transport sectors, emissions were provided for the 4 marker scenarios A1, A2, B1, and B2. In Act.3 we have decided to start with the high emission scenario A1 and the low emission scenario B1. In the case of aviation the “A1i” (moderate to good fuel efficiency, moderate NOx improvements) and the “B1” (good fuel efficiency improvement, good NOx improvements) versions will be used.

* man month numbers are given as: planned for month 25-42 (spent in month 25-36)

As was done in the year 2000/2003 simulations, we will reduce one transport sector at a time also in the future runs (i.e. three runs with 5% reduction in aviation, road, shipping, respectively), in addition to a reference future run with all emissions included.

For well-mixed GHG it has been decided to use the SRES recommendations. So far, UiO have provided tables for the A1b scenario. Source data for sea surface temperatures has still to be decided. Chlorine and bromine levels from WMO2006 will be used in those models that have stratospheric chemistry. Tables were provided by UiO.

All necessary simulations will be done during spring/summer 2008.

Deliverables

D3.3.1.2	Future chemical perturbation fields from the CNRS-LSCE model (month 36, CNRS-LSCE, now planned month 42)
D3.3.1.3	Future chemical perturbation fields from the KNMI model (month 36, KNMI, now planned month 42)
D3.3.1.4	Future chemical perturbation fields from the MPICHEM model (month 42, MPICHEM) May be joint with DLR-IPA (D3.3.1.8).
D3.3.1.5	Future chemical perturbation fields from the UCI model (month 36, UCI, now planned month 42)
D3.3.1.6	Future chemical perturbation fields from the UCAM-DCHEM model (month 36, UCAM-DCHEM, now planned month 42)
D3.3.1.7	Future chemical perturbation fields from the UIO model (month 36, UIO, now planned month 42)
D3.3.1.8	Future chemical perturbation fields from the DLR model (month 42, DLR)
D3.3.1.9	Report with estimate of the future chemical impact (month 46, UIO)

Milestones

M3.3.1.1	Detailed description of model simulations to be performed accomplished (month 9, UIO, done)
M3.3.1.2	Results from simulations future impact from CTMs available (month 36, UIO, now planned month 42)
M3.3.1.3	All results from simulations of future impact available, including those from CCMs (month 42, UIO)

WP 3.3.2 Impact of future climate change (Grewe, DLR-IPA)

WP number	3.3.2	Period month 25-42		Start date :	25	End date	48
Participant id	DLR-IPA	CNRS-LSCE	MPICHEM	UiO			
Person-months*	9 (2.4)	9 (0)	6 (0)	5 (0)			

Objectives

O3.3.2.1 To estimate the effect of atmospheric chemistry-climate feedbacks on the future impact of emissions from transport (M3.3.2.3)

Description of work

Significant changes in climate parameters such as H2O, temperature, and dynamics (transport of stratospheric ozone) are expected to occur in the future global atmosphere. These changes will affect the oxidation process and thereby the atmospheric concentrations of climate gases such as ozone and methane. In a first step existing global ACM simulations will be analysed with regard to the impact of climate variability and of chemistry-climate feed-backs on traffic induced ozone changes.

In a second step each participating group will perform 4 simulations (the first two of which are in common with WP3.3.1, i.e. A and BR)

A: A simulation with meteorology/climate for 2000 and all emissions for 2050.

BR: A simulation with meteorology/climate for 2000 and all emissions for 2050 except for a reduction of the emissions of one representative sector (e.g. road traffic) by -5%.

C: Same as A but with meteorology/climate for 2050.

DR: Same as BR but with meteorology/climate for 2050

Description of progress

The set-up for the model simulations has been discussed and described in deliverable D3.3.2.1 that was submitted by DLR in month 31. After presentation of the future scenarios at the General Assembly in Budapest in February 2008 it has been decided to modify the BR simulation into a simulation where all transport emissions are reduced, instead of only road traffic. The reason is that the future scenarios for road traffic take into account quite a lot of expected mitigation measures and therefore are rather low for 2050 and later. The runs have not started yet, for the same reason as in WP3.3.1: the gridded future emission data were delivered rather late by Act. 1 and still require further evaluation and preprocessing, which are currently underway. It is still planned to finish the simulations by month 42.

Deliverables

D3.3.2.1 Document describing the setup of the chemistry-climate feedback model experiments (month 28, DLR-IPA, submitted month 31)

D3.3.2.2 Future chemical perturbation fields including climate feedbacks from the DLR-IPA model (month 42, DLR-IPA)

D3.3.2.3 Future chemical perturbation fields including climate feedbacks from the CNRS-LSCE model (month 42, CNRS-LSCE)

D3.3.2.4 Future chemical perturbation fields including climate feedbacks from the MPICHEM model (month 42, MPICHEM). May be joint with DLR-IPA (D3.3.2.1).

D3.3.2.5 Future chemical perturbation fields including climate feedbacks from the UIO model (month 42, UIO)

* man month numbers are given as: planned for month 25-42 (spent in month 25-36)

Milestones	
M3.3.2.1	Design of model experiments available (month 28, DLR-IPA, done)
M3.3.2.2	Model simulations ready (month 42, DLR-IPA)

WP 3.3.3 Possible mitigation strategies (Isaksen, UIO)

WP number	3.3.3	Period month 25-42	Start date :	19	End date	40
Participant id	CNRS-LSCE	KNMI	MPICHEM	UIO	UCAM-DCHEM	
Person-months[*]	2 (0)	2 (0)	1 (0)	2 (1)	1 (0)	

Objective

O3.3.3.1 To investigate options for reducing the impact of individual transport modes and regions

Description of work

Model runs with the global scale CTMs will be performed to estimate different options for reduced climate and air quality impact. The model runs will look at contributions from individual modes of transport and from different regions. Analysis and recommendations for combination of options that efficiently reduce the impact of emissions will be made. Options will include implementation of catalytic converters in regions where they are not implemented, introduction of alternative energy (hydrogen economy), cleaner fuel (ship traffic) and change in traffic pattern and cargo transport. The studies will include climate compounds and pollutants like CO2, O3, CH4 (through changes in its lifetime) and particles (sulphate, organic carbon aerosols).

Target years are 2050 and 2100 Selection of scenarios for these studies will be performed in close collaboration with Activity 1. Results from the analysis will be provided to Activity 7 dealing with impact assessment.

Compared to the original DoW WP3.3.3 has been slightly shifted forward in time, while WP3.3.1 has been shifted backward in time – in order to allow Activity 1 to provide emission input data in time.

Description of progress

D3.3.3.1 (description of model setup for calculating effects of mitigation options) was submitted in month 30. As the provision of mitigation scenarios of emissions is not included in the work plan for Activity 1, alternative solutions had to be found. The plan is now to run the "B1 ACARE" scenario, provided by Activity 1, for aviation as a mitigation scenario. B1 ACARE has the lowest emissions due to the implementation of the ACARE targets for fuel and NOx: this can be described as a mitigation scenario as the fuel efficiency improvements are most likely to be implemented due to concern over climate change. B1 ACARE can be seen as a very optimistic but feasible scenario. (ACARE=Advisory Council for Aeronautics Research in Europe has formulated environmental targets for the industry, published in their Strategic Research Agenda, http://www.acare4europe.org/docs/ASD-exec%20sum-2nd-final-171104-out-asd.pdf).

For road it was originally envisaged to create a hydrogen scenario that assumes increased use of hydrogen on top of scenario B1. During the third year of Quantify the scenario has been discussed intensively by Activity 1 and Activity 3 members. Details were described in the delivery report D3.3.3.1. As emissions of short-lived species (ozone precursors) will be largely eliminated by the year 2050, the scenario would focuss mostly on the reduction of CO2.

However, at the third annual meeting in Budapest, it was found that the future scenario for road traffic produced by Activity 1 already looks more or less like a mitigation scenario (having taken into account significant planned reductions), while for the future shipping and aviation scenarios this is not the case. Therefore, it was decided to create a 'policy failure' scenario for road emissions. It is likely that this scenario will replace the hydrogen scenario that was previously planned.

[*] man month numbers are given as: planned for month 25-42 (spent in month 25-36)

Deliverables	
D3.3.3.1	Description of model setup for calculating effects of mitigation options (month 24, UIO, submitted month 30)
D3.3.3.2	Datasets from the CNRS-LSCE model with chemical perturbations for different mitigation options (month 36, CNRS-LSCE, will be shifted to month 45)
D3.3.3.3	Datasets from the KNMI model with chemical perturbations for different mitigation options (month 36, KNMI, will be shifted to month 45)
D3.3.3.4	Datasets from the MPICHEM model with chemical perturbations for different mitigation options (month 36, MPICHEM, will be cancelled)
D3.3.3.5	Datasets from the UCAM-DCHEM model with chemical perturbations for different mitigation options (month 36, UCAM-DCHEM, will be shifted to month 45)
D3.3.3.6	Datasets from the UIO model with chemical perturbations for different mitigation options (month 36, UIO, will be shifted to month 45)
D3.3.3.7	Datasets from the UCI model with chemical perturbations for different mitigation options (month 36, UCI, will be shifted to month 45)
D3.3.3.8	Report on mitigation strategies (month 40, UIO, will be shifted to month 48)

Milestones	
M3.3.3.1	Selection done of reduction options to be investigated (month 30, UIO)
M3.3.3.2	Model simulations ready; provision of output data to Activity 7 accomplished (month 46 UIO)