SPECIAL PROJECT FINAL REPORT

All the following mandatory information needs to be provided.

Project Title:	Inline chemistry for tropospheric and stratospheric trace gases in IFS
Computer Project Account:	SPNLMACC
Start Year - End Year :	2015- 2017
Principal Investigator(s)	V. Huijnen
Affiliation/Address:	Royal Netherlands Meteorological Institute (KNMI) PO Box 201, 3730 AE De Bilt The Netherlands
Other Researchers (Name/Affiliation):	

The following should cover the entire project duration.

Summary of project objectives
(10 lines max)

Composition-IFS where CB05 tropospheric chemistry is integral part of the IFS code, allows unprecedented couplings between chemical tracer fields with aerosol and meteorological aspects. The aims of the current special project are (i) to extend the existing chemical scheme, by introducing stratospheric chemistry from the BASCOE scheme, (ii) to optimize the solver, through the use of a pre-processor, and (iii) to explore new interactions specifically related to the new aerosol model. This aims to support the work performed in the MACC-III and scientific aspects related to the more operational tasks done in the framework of the Copernicus Atmospheric Monitoring Service.

Summary of problems encountered
(If you encountered any problems of a more technical nature, please describe them here.)

No significant problems have occurred

Experience with the Special Project framework
(Please let us know about your experience with administrative aspects like the application procedure, progress reporting etc.)

It is sometimes unclear which type of reporting is requested by whom (national representatives and/or ECMWF directly), and when reports are due. More consistent reminder emails from various stakeholders (and a little coordination) could be beneficial. Otherwise all is fine.

Summary of results
(This section should comprise up to 10 pages and can be replaced by a short summary plus an existing scientific report on the project.)

The results of this special project have been split into four sections: 1. Development and evaluation of stratospheric chemistry in IFS; 2. Application of automated code generator for the chemistry solver; 3. Chemistry-aerosol interaction and 4. Application of Composition-IFS for a case study.

1. Stratospheric chemistry in IFS

Over the course of the whole project period, efforts have been made on the development and evaluation of the stratospheric chemistry in the IFS. This has resulted in a publication (Huijnen et al., 2016a). The approach where tropospheric and stratospheric chemistry have been computed in their respective domains, referred to as 'C-IFS-CB05-BASCOE', has been compared to an approach which includes all reactions throughout the atmosphere, 'C-IFS-Atmos'. Also attention has been given to the detection and prevention of drifts in composition fields, for multi-annual model experiments.

Huijnen et al. (2016a) documents the performance of stratospheric ozone in C-IFS-CB05-BASCOE and C-IFS-Atmos against observations from ozone sondes for 2009 and 2010. An example of this evaluation is illustrated in Fig. 1, which shows the evolution of stratospheric O_3 against observations at the Syowa (Antarctica) station. It clearly shows the ability of the C-IFS versions that have stratospheric chemistry to capture the ozone hole. The relatively marginal difference between C-IFS-CB05-BASCOE and C-IFS-Atmos indicates that our approach where tropospheric and

stratospheric chemistry are only switched on in troposphere and stratosphere, respectively, leads to very similar results as a setup where the full reaction schemes for both troposphere and stratosphere are retained throughout the atmosphere (C-IFS-Atmos). A closer look at the difference in trace gas profiles between C-IFS-CB05-BASCOE and C-IFS-Atmos (Figure 3) gives further confidence in this approach. Furthermore, C-IFS-CB05-BASCOE appears computationally significantly less computationally expensive compared to the approach in C-IFS-Atmos. Assessment of trends in O_3 total columns for multi-year simulations suggest in fact that biases are present within C-IFS, but become essentially stabilized after about 1 year simulation time (Figure 3). This system has been delivered to CAMS, and is further evaluated and developed in the framework of the CAMS tender on global reactive gases.
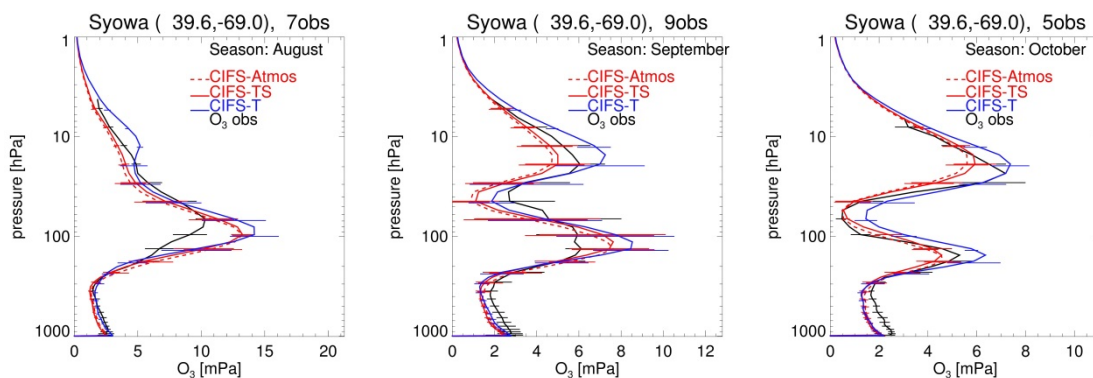

Figure 1. Evaluation of stratospheric ozone against WOUDC ozone sondes at Syowa station during September-November 2009. Black: observations, Red solid: C-IFS-CB05-BASCOE, red dashed: C-IFS-Atmos, blue solid: C-IFS-Cariolle.
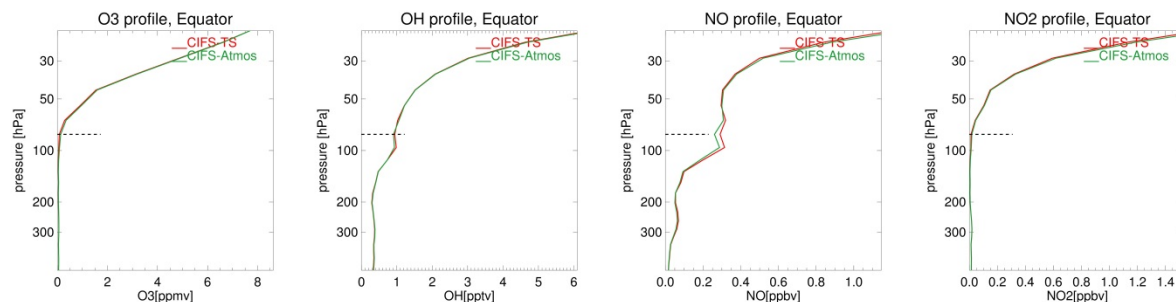

Figure 2. Instantaneous profiles of O_3, OH, NO and NO_2 at 0°N, 180°W, on 1 October 2008, 0 UTC, for runs C-IFS-CB05-BASCOE (red) and C-IFS-Atmos (green). The dashed line denotes the chemical tropopause level associated with the switch in the chemistry mechanism.
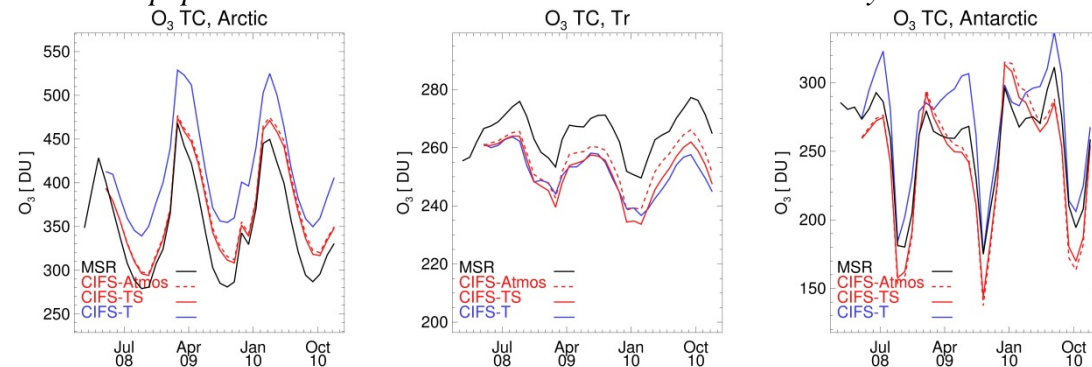

Figure 3. Evaluation of C-IFS-CB05-BASCOE (red, solid), C-IFS-Atmos (red, dashes) and CIFS-Cariolle (blue) monthly mean O_3 total columns in Dobson Units against the Multi-Sensor Reanalysis for the Arctic (90°N-70°N), tropics (30°N-30°S) and Antarctica (70°S-90°S).

2. Use and optimization of pre-processor to provide a solver for atmospheric chemistry

Over the course of this special project technical activities have been performed to assess the replacement of the baseline Euler Backward Integration Method for solving tropospheric chemistry with a KPP-based Rosenbrock solver, which simplifies chemistry version management and may provide improved accuracy. However, application of the Rosenbrock solver appeared significantly slower than the standard EBI solver, while model results are also not identical. Also, KPP does not follow IFS coding conventions, and efforts are needed to automatize such code generation. Finally, stratospheric chemistry showed spurious drifts which in part could be attributed to KPP-solver settings (convergence). Technical and scientific efforts have been pursued on these various aspects, as documented in CAMS deliverable report D42.1.5.4 (2018), which has been aligned with the activity in this special project. From this, we highlight here the conclusions reached.

Increased accuracy for stratospheric chemistry runs
An assessment of differences in vertical profiles after a 1.5 year IFS(CB05BASCOE) run with focus on stratospheric chemistry are shown in Figure 4. It illustrates a significant change in model profiles when stricter tolerance levels are adopted of 0.005, compared to a baseline relative tolerance in BASCOE-CTM of 0.1, overall leading to results that are better in line with an independent BASCOE-CTM model experiment. Also significantly improved performance against observations has been reached (not shown here).
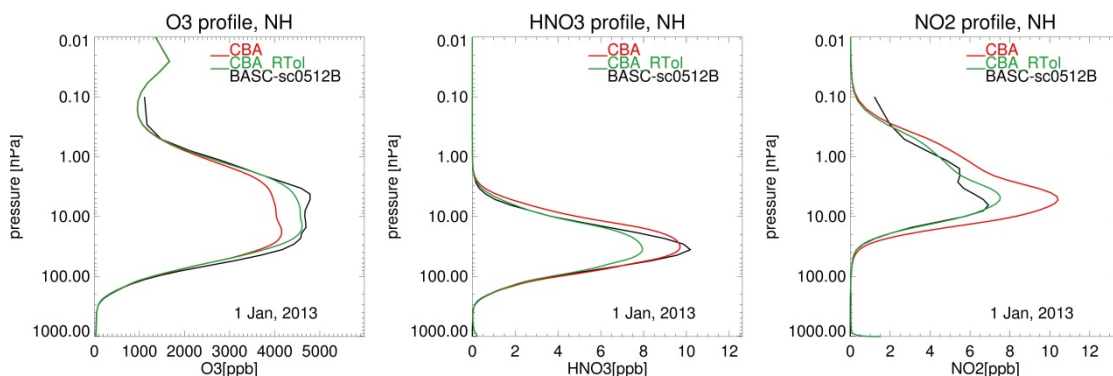

Figure 4. Illustration of differences in instantaneous, zonal mean vertical profile over the NH-extra tropics (30N-90N) for 1 January 2013 for runs CBA and CBA_RTOL, which applies a stricter solver convergence criterion. Black line: BASCOE-CTM results.

Code optimization
The KP4 code for the LU Decomposition ('KppDecomp')has been implemented for CB05 and BASCOE schemes, Jöckel et al. (2010). In particular, in nested loops for the computation of LU-decomposition of the (sparce) Jacobian contain in the original KPP implementation integer arrays for indirect addressing of arrays. Since the indices are known a-priori, depending on the specific kinetic mechanism, the loops can be easily replaced by a sequence of operations without the need of indirect addressing. Application of the KP4 option was shown to lead to an increase of overall runtime efficiency by ~10% in the 60-level model version.

Application of KPP for tropospheric chemistry
Using the KPP-based Rosenbrock solver for tropospheric chemistry was shown to lead to a decrease in O_3 together with an increase in carbon monoxide during summer periods (both for the NH- and SH-extra tropics). In particular a marked decrease of ~20-30% is visible for tropospheric ozone over the outflow of the Arabian peninsula during July. This appears associated to a reduced NOx lifetime over the region, due to larger efficiency of heterogeneous loss of N_2O_5 over aerosol particles. Evaluation of the model against OMI observations of tropospheric NO_2, Fig 5. shows a significant over-estimate over the Arabian peninsula in the reference simulation which is reduced in run which applies the Rosenbrock solver.
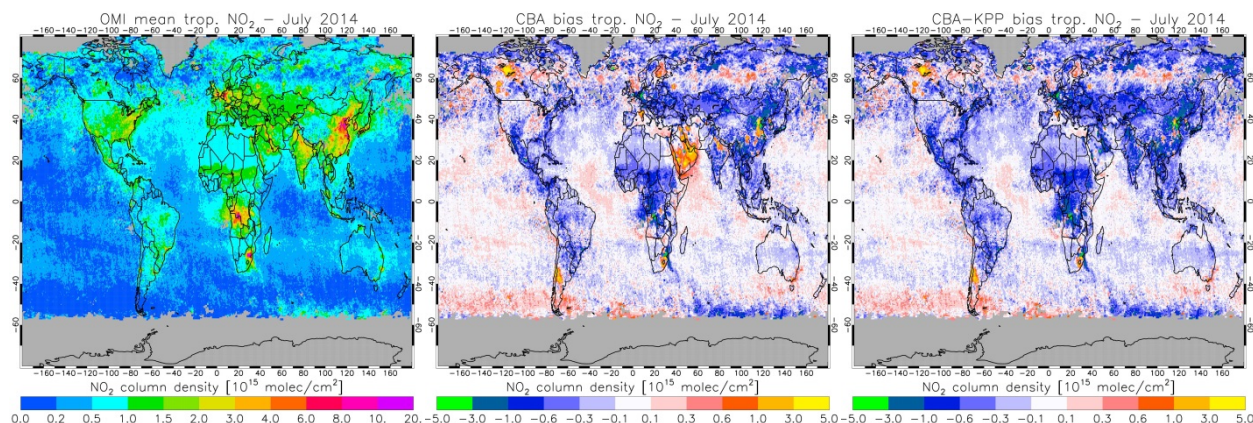
Figure 5. Evaluation of tropospheric NO₂ against OMI for July 2014. Left: Observations. Middle: bias for the run using EBI solver. Right: bias for run using Rosenbrock-solver.

3. Chemistry-aerosol interaction

In the framework of this project we focussed on the development of an efficient secondary organic aerosol (SOA) module. This involves chemical production of gas-phase anthropogenic and biogenic SOA precursors (SOG) and a module to describe SOG-SOA equilibrium, depending on assumed volatility. The following results have been achieved over the course of this project.

The modelling of SOA precursors have been matured by introducing tracers for aromatics (xylene and toluene), and corresponding emission sources, as well as ageing of SOG, making them larger and less volatile, Tsimpidi et al (GMD 2014). We keep track of three individual SOA tracers:
(1) biogenic SOA, produced from terpene and isoprene oxidation with OH, O_3 and NO_3
(2) two classes with different volatility bins of 'anthropogenic' SOA, produced from OLE, PAR and C_3H_6 and aromatics as oxidized from OH, O_3 and NO_3.

First tests have been performed with this scheme, see Table 1 for annual budgets, and Figure 6 illustrating monthly average surface concentrations. Compared to literature (Hodzic et al., ACP 2016), the total budgets appear in reasonable agreement with their more detailed approach, although latest findings, and observational evidence, suggest missing SOA sources, as well as shorter SOA lifetimes.

Whereas these aerosols were initially described within framework of chemical tracers, they are now officially part of the CAMS aerosol package, establishing a firm link between gas-phase chemistry and aerosol, and hence allowing a further evaluation of, e.g., their impacts on AOD, which is subject of future work.

Table 1. Annual budgets of the two SOG/SOA types as implemented in IFS

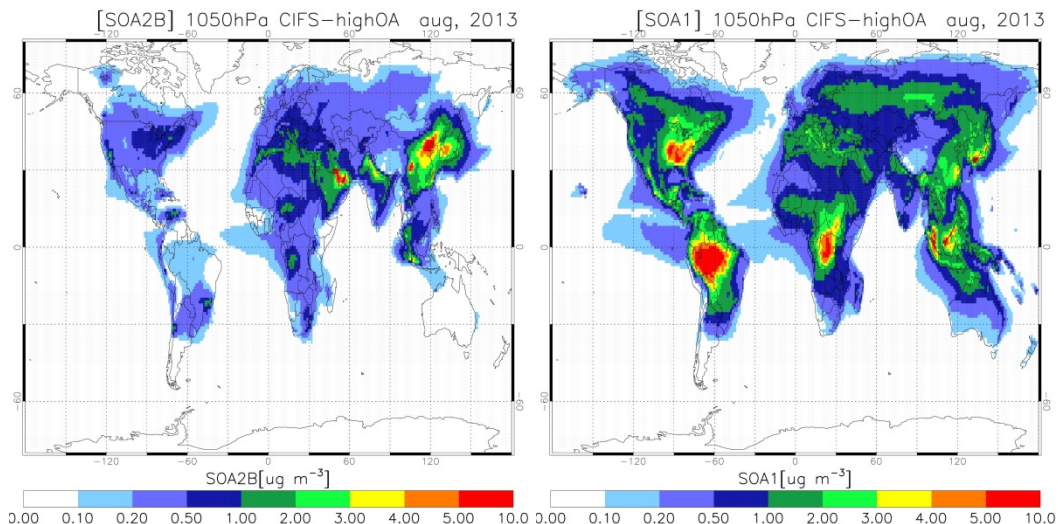| This work	Gas prod [Tg yr⁻¹]	Gas burden [Tg]	Gas drywetdep [Tg yr⁻¹]	Particle prod. [Tg yr⁻¹]	Particle wdep [Tg yr⁻¹]	Particle burden [Tg]	Particle lifetime [days]
Anthro	22	0.17	10.2	15.5	15.5	0.35	8.3
Bio	38.6	0.037	5.0	33.7	33.7	0.71	7.7

Figure 6: Illustration of surface concentrations of anthropogenic (left) and biogenic (right) SOA averaged for August 2013.

4. Evaluation of current ability of the system for a very large fire event

In response to the large fires over Indonesia during the El Nino draught conditions in September-October 2015, we decided to quickly perform a C-IFS-based study has been setup quickly where the GFAS fire emissions have been evaluated and optimized in order to estimate the total CO_2 emissions resulting from the peatland fires, to exploit the ability of the system developed within CAMS. Although seasonal fires are a frequent occurrence in the human modified landscapes found in Indonesia, the extent of the 2015 fires was greatly inflated by an extended drought period associated with a strong El Niño. Our methodology is based on a hand-guided inversion methodology for C-IFS simulations of CO, constrained with MOPITT, where we use actually observed in-situ emission ratios of CO_2 and CH_4 to CO. Based on that, we estimated that total carbon released by the fires during Sept-Oct 2015 was 227 ± 67 Tg C, of which 83% is in the form of CO_2 (692 Tg CO_2), 16% CO (84 Tg CO), and 1% CH_4 (3.2 Tg CH_4). We estimate carbon emissions from the 2015 fires to be the largest seen in maritime southeast Asia since those associated with the record breaking El Niño of 1997. Figure 7 shows a map with the spatial distribution of our optimized CO_2 emissions over the region. This study has led to a well-cited publication (Huijnen et al., 2016b).
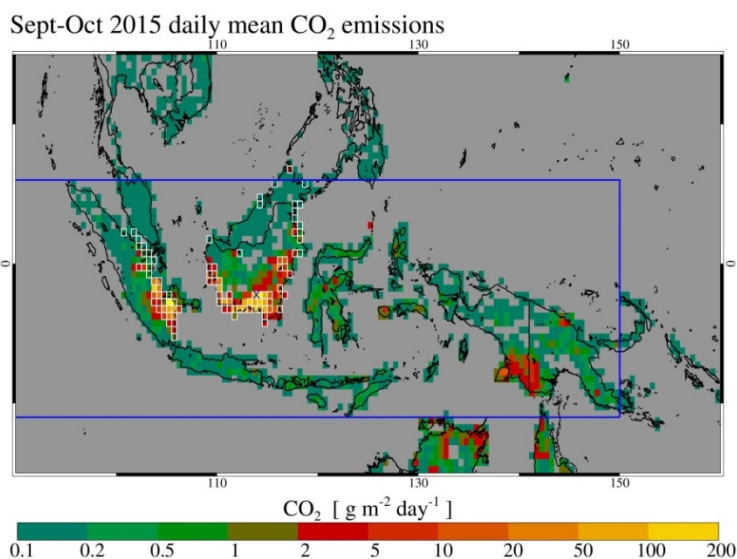

Figure 7. Daily mean CO_2 emissions from peat and vegetation fires burning across maritime southeast Asia in Sept-Oct 2015, presented in $0.5° \times 0.5°$ grid cells. Cells containing peat soils according to landcover data used in GFASv1.2 are outlined in white.

List of publications/reports from the project with complete references

Eskes, H., Huijnen, V., Arola, A., Benedictow, A., Blechschmidt, A.-M., Botek, E., Boucher, O., Bouarar, I., Chabrillat, S., Cuevas, E., Engelen, R., Flentje, H., Gaudel, A., Griesfeller, J., Jones, L., Kapsomenakis, J., Katragkou, E., Kinne, S., Langerock, B., Razinger, M., Richter, A., Schultz, M., Schulz, M., Sudarchikova, N., Thouret, V., Vrekoussis, M., Wagner, A., and Zerefos, C.: Validation of reactive gases and aerosols in the MACC global analysis and forecast system, Geosci. Model Dev., 8, 3523-3543, doi:10.5194/gmd-8-3523-2015, 2015.

Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M., Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou, E., Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A.: Tropospheric chemistry in the Integrated Forecasting System of ECMWF, Geosci. Model Dev., 8, 975-1003, doi:10.5194/gmd-8-975-2015, 2015.

Flemming, J., Benedetti, A., Inness, A., Engelen, R. J., Jones, L., Huijnen, V., Remy, S., Parrington, M., Suttie, M., Bozzo, A., Peuch, V.-H., Akritidis, D., and Katragkou, E.: The CAMS interim Reanalysis of Carbon Monoxide, Ozone and Aerosol for 2003–2015, Atmos. Chem. Phys., 17, 1945-1983, https://doi.org/10.5194/acp-17-1945-2017, 2017.

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Huijnen, V., M. J. Wooster, J. W. Kaiser, D. L. A. Gaveau, J. Flemming, M. Parrington, A. Inness, D. Murdiyarso, B. Main and M. van Weele. Fire carbon emissions over maritime southeast Asia in 2015 largest since 1997. Sci. Rep. 6, 26886; doi: 10.1038/srep26886, 2016b.

Inness, A., Blechschmidt, A.-M., Bouarar, I., Chabrillat, S., Crepulja, M., Engelen, R. J., Eskes, H., Flemming, J., Gaudel, A., Hendrick, F., Huijnen, V., Jones, L., Kapsomenakis, J., Katragkou, E., Keppens, A., Langerock, B., de Mazière, M., Melas, D., Parrington, M., Peuch, V. H., Razinger, M., Richter, A., Schultz, M. G., Suttie, M., Thouret, V., Vrekoussis, M., Wagner, A., and Zerefos, C.: Data assimilation of satellite-retrieved ozone, carbon monoxide and nitrogen dioxide with ECMWF's Composition-IFS, Atmos. Chem. Phys., 15, 5275-5303, doi:10.5194/acp-15-5275-2015, 2015.

Inness, A., Benedetti, A., Flemming, J., Huijnen, V., Kaiser, J. W., Parrington, M., and Remy, S.: The ENSO signal in atmospheric composition fields: emission-driven versus dynamically induced changes, Atmos. Chem. Phys., 15, 9083-9097, doi:10.5194/acp-15-9083-2015, 2015.

Wooster, M.J., Gaveau, D.L.A., Salim, M.A., Zhang, T., Xu, W., Green, D.C., Huijnen, V., Murdiyarso, D., Gunawan, D., Borchard, N., Schirrmann, M., Main, B., Sepriando, A. New tropical peatland gas and particulate emissions factors indicate 2015 Indonesian fires released far more particulate matter (but Less Methane) than current inventories imply. Remote Sensing, 10 (4), 2018.

Future plans

(Please let us know of any imminent plans regarding a continuation of this research activity, in particular if they are linked to another/new Special Project.)

A follow-up special project has been requested (SPNLMACC) which aims to cover resources needed for scientific spinoffs from the efforts done in the framework of the Copernicus Atmosphere Monitoring Service (CAMS) tender on reactive gases modelling and assimilation. Topics under consideration are: 1) a further integration of tropospheric and stratospheric chemistry; 2) further integration of chemistry and aerosol subjects, particularly stratospheric aerosol and secondary organic aerosol; 3) Re-evaluate specific case studies.