

SPECIAL PROJECT PROGRESS REPORT

Progress Reports should be 2 to 10 pages in length, depending on importance of the project. All the following mandatory information needs to be provided.

Reporting year 2018

Project Title: Chemistry and aerosol processes modelling within IFS in extension to CAMS activities

Computer Project Account: SPNLMACC

Principal Investigator(s): Dr. V. Huijnen

Affiliation: Royal Netherlands Meteorological Institute (KNMI)

Name of ECMWF scientist(s) collaborating to the project
(if applicable) J. Flemming

Start date of the project: 2018

Expected end date: 2020

Computer resources allocated/used for the current year and the previous one (if applicable)

Please answer for all project resources

		Previous year		Current year	
		Allocated	Used	Allocated	Used
High Performance Computing Facility	(units)	1400k	1100k	1500k	250k
Data storage capacity	(Gbytes)				

Summary of project objectives

(10 lines max)

Within the Copernicus Atmosphere Monitoring Service (CAMS) modules for tropospheric and stratospheric chemistry, integrated in ECMWF's Integrated Forecasting System (IFS), are maintained and further improved within the CAMS tender on reactive gases modelling and assimilation, but does not cover extended scientific spinoff's from this work. This special project aims to provide us with in the computer resources needed for this. 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, particularly the 2015 Indonesian fires.

Summary of problems encountered (if any)

(20 lines max)

No significant problems have occurred

Summary of results of the current year (from July of previous year to June of current year)

1. Integration and extension of tropospheric and stratospheric chemistry

In the framework of CAMS_42 multi-annual model experiments with stratospheric chemistry have been executed, which revealed drifts in composition fields, e.g. stratospheric NO_x , HNO_3 and chlorine containing trace gases. Efforts to prevent these drifts have been given priority in this year, before further steps could be made. This resulted indeed in much more robust chemistry configurations, although biases still exist in the BASCOE-description of stratospheric chemistry.

An analysis of stratospheric photolysis rates showed that it is close to the tropospheric parameterization if cloud effects are switched off (Figure 1). A test experiment where photolysis of NO_2 for the troposphere has been extended into the stratosphere revealed slightly reduced stratospheric day-time NO_2 concentrations. It was shown to improve slightly the comparison to OMI NO_2 retrievals, but did only show marginal impact on O_3 (not shown here).

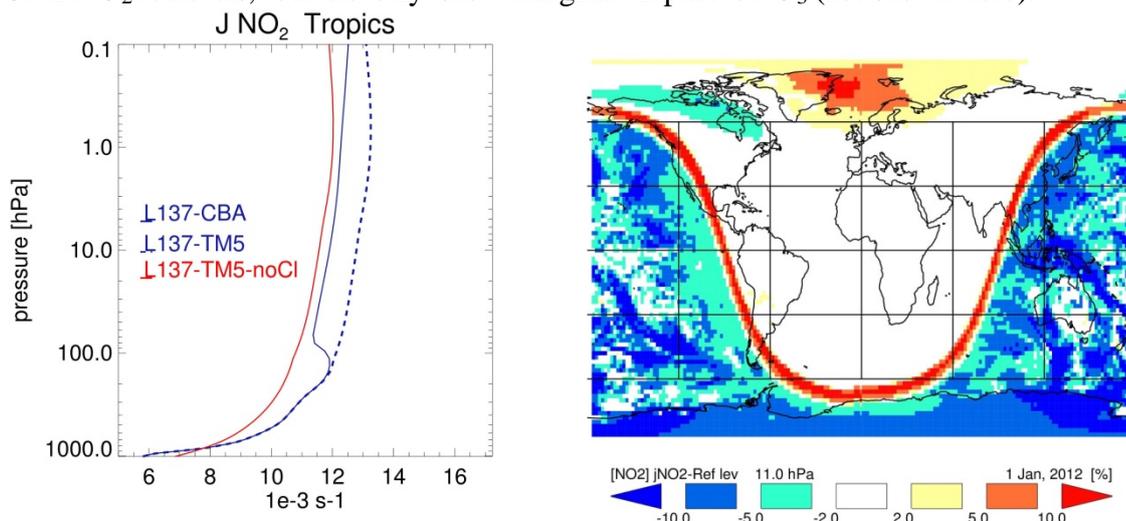


Figure 1: Left: Intercomparison of average NO_2 photolysis rate as computed using the stratospheric parameterization (blue, solid above 100 hPa), and tropospheric parameterization (blue dashed), as well as the tropospheric parameterization but excluding cloud effects (red). In stratosphere this is close to the stratospheric parameterization (which also doesn't take cloud scattering into account). Right: percentual change in instantaneous NO_2 concentrations at 10 hPa when applying higher tropospheric $j\text{NO}_2$ parameterization.

2. Secondary organic aerosol

A new module to describe efficiently the secondary organic aerosol (SOA) production linked to chemistry precursors has been developed. This involves reversible formation of gas-phase anthropogenic and biogenic SOA precursors (SOG), where gas-phase VOC's are assumed in equilibrium with aerosol, depending on the volatility of the respective classes of molecules. Since last year the following activities have been pursued.

Compared to the status of previous year, anthropogenic emission sources from aromatics (xylene, toluene) have now been included, as well as ageing of SOG, making them larger and less volatile, Tsimpidi et al (2014). Also we now keep track of three individual SOA tracers:

- (1) biogenic SOA, produced from terpene and isoprene oxidation with OH, O₃ and NO₃
- (2) two classes anthropogenic SOA produced from OLE, PAR and C₃H₆, and aromatics oxidation with OH, O₃ and NO₃.

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

Table 1 lists their global production and loss budgets.

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

Compared to literature (Hodzic et al., 2016), these totals appear in reasonable agreement with their more detailed approach, although latest findings, and observational evidence, suggests missing SOA sources, as well as shorter SOA lifetimes. To resolve this, we opt for a hybrid approach where also direct SOA emissions are introduced following the pattern of anthropogenic CO emissions, and optimize the SOA burdens in comparison to station measurements as well as observations of AOD.

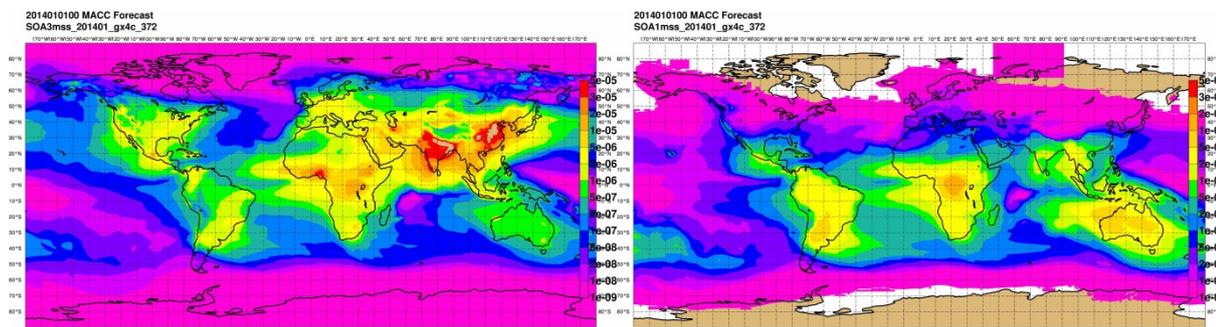


Figure 2. Illustration of monthly mean anthropogenic (left) and biogenic (right) SOA total columns (kg m^{-2}) for January 2014.

3. Re-evaluation of case studies

In support to a study to provide best-estimates of CO (and CO₂) emissions originating from the 2015 fires over Indonesia we based on a 4D-Var inversion system (Banda et al., submitted), we ran several IFS model experiments with different emission estimates. From this, we assessed the impact on OH, which defines the lifetime of CO, and is hence an important, yet uncertain, parameter in inversion studies. Figure 3 shows the resulting average CO columns for 1-10 Oct 2015, and their respective biases compared to MOPITT. It illustrates significant sensitivity to assumed emission factors, particularly due to the uncertain ratio between CO and NO_x emissions. The sensitivity of inverted CO emissions to assumed OH is used in the Banda et al. manuscript.

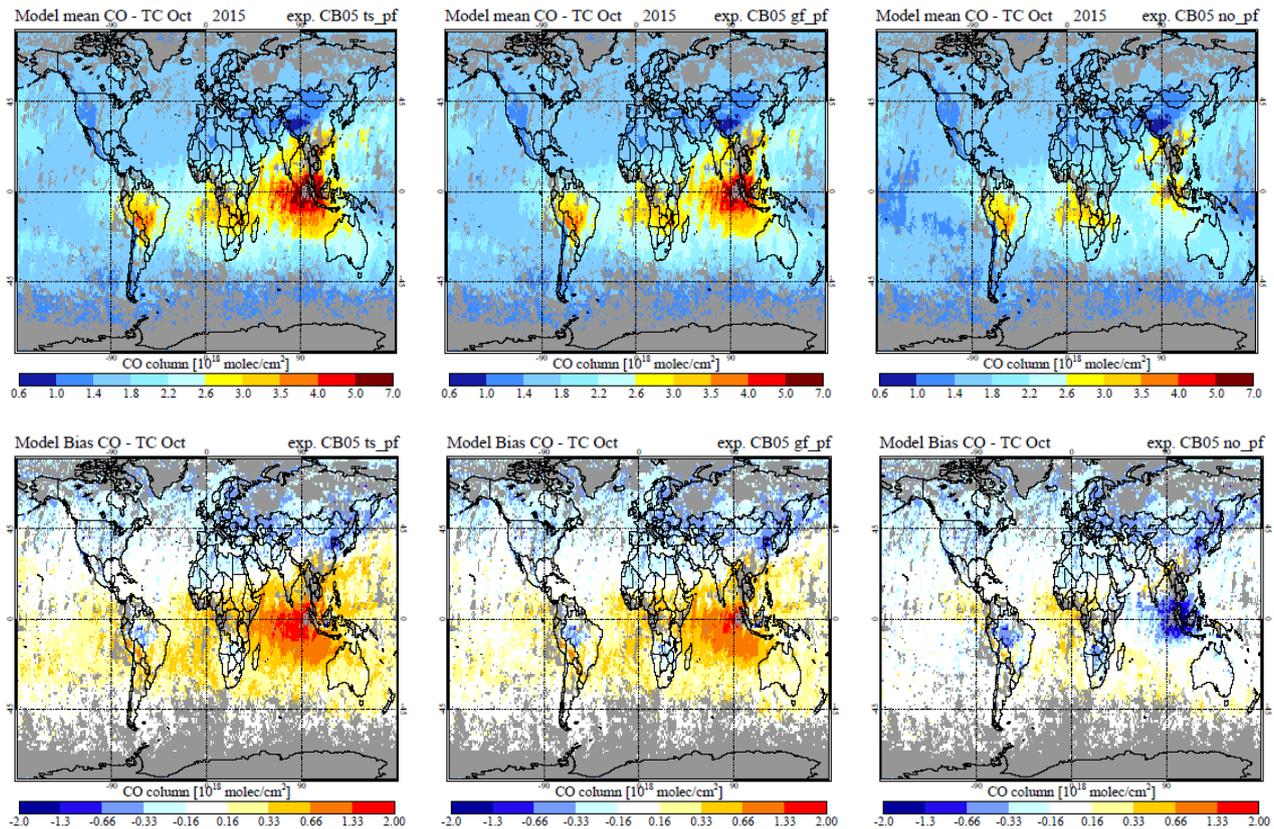


Figure 3. Top row: CO total columns during 1-10 Oct 2015 from sensitivity experiments with IFS(CB05) using various emission scenarios. Left: emission factors from Stockwell et al., middle: GFED emission factors. Right: No peat fire emissions included. Bottom row: Biases against corresponding MOPITT observations.

List of publications/reports from the project with complete references

- Banda et al., Monitoring emissions from the 2015 Indonesian fires using CO satellite data, submitted to Phil. Trans. Royal Soc. B (2018)
- 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.
- Huijnen, V., Flemming, J., Chabrillat, S., Errera, Q., Christophe, Y., Blechschmidt, A.-M., Richter, A., and Eskes, H.: C-IFS-CB05-BASCOE: stratospheric chemistry in the Integrated Forecasting System of ECMWF, *Geosci. Model Dev.*, 9, 3071-3091, doi:10.5194/gmd-9-3071-2016, 2016a.
- 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.

Summary of plans for the continuation of the project

(10 lines max)

In the second year of this special project we plan to evaluate more closely the stratospheric composition, with focus on CH₄ and H₂O. Also further develop the implementation of the inorganic and organic aerosol coupling with chemistry precursors, in particular for the organic aerosol further work is needed in terms of model tuning and evaluation. Also a first integration with the GLOMAP model, which is expected to become officially available in the upcoming cycle, is foreseen.