TECHNICAL MEMORANDUM

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Atmospheric Composition priority developments for Numerical Weather Prediction

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Abstract

One of the ECMWF's strategic goals for 2025 is to develop an integrated global model of the Earth system to produce forecasts with increasing fidelity up to a year ahead. This will be achieved by incorporating an increased level of complexity of physical and chemical processes and of the interactions between the different Earth's components into the model. Atmospheric composition (AC) has the potential to be one of the sources of predictability at different time scales. Development activities at ECMWF have built a capacity to simulate and assimilate a variety of AC species in the Integrated Forecasting System (IFS). Today this capacity is at the core of the Copernicus Atmosphere Monitoring Service (CAMS), but the complexity of many AC modules in the IFS often makes them computationally unaffordable at the resolutions used in numerical weather prediction (NWP). Only stratospheric ozone is a prognostic variable in NWP applications, and it is included interactively in the radiative transfer model used for radiance assimilation. The radiation scheme still relies on climatologies of aerosols, ozone and trace gases, although considerable collaborative effort has been made in the last few years to use the CAMS products to improve the realism of these AC climatologies and of the related AC-weather feedbacks.

This document recommends AC priority developments for NWP that has the potential to further improve the forecasts from days to seasons ahead. These are considered for testing and possibly operational implementation by 2022. The proposal is to improve the representation of AC in NWP to the level of complexity and coupling beneficial for NWP by leveraging the CAMS developments wherever possible. Hence, the focus is on ozone, aerosols, and CO_2. In addition to modelling and data assimilation aspects, attention will also be paid to code structure and code efficiency, and, provided a higher level of coupling between AC and meteorology is introduced, to how the model performance evaluation process might need to be adapted in the future.

1 Introduction

It is recognized that atmospheric composition (AC) species, such as ozone, aerosols and greenhouse gases, affect air quality, weather, and climate. Changes in their concentrations can alter the balance between the amount of incoming solar radiation reaching the surface of the Earth and that of the outgoing radiation emitted at the surface to space. Modifying such a balance has consequences on the atmospheric circulation both globally and locally over time scales that can range from days to years. For instance, Charlton et al (2004) showed that the modelling and assimilation of stratospheric ozone in atmospheric general circulation models can lead to a better representation of the stratospheric dynamics and in turn influence the tropospheric weather. Aerosols have also received attention by the NWP (acronyms not defined in the text are available in Appendix D) community, as outlined in studies like Rodwell and Jung (2008), Reale et al. (2011), and Mulcahy et al (2014). The WMO has also initiated a series of studies to understand better the impact of AC in NWP (Baklanov et al, 2017).

Because of this ability to interact with the meteorology and to affect both weather and climate, AC has received increasing attention by the weather and climate communities especially over the last two decades. This has resulted in some level of modelling complexity of atmospheric composition in NWP and climate models. The availability of satellite observations of AC constituents with increasingly good quality has also facilitated the development of global data assimilation systems able to optimally combine these observations with the meteorological data to estimate the state of the atmosphere (Hollingsworth et al, 2008).

ECMWF started AC developments in the late 1990s when a stratospheric ozone model and data assimilation system were developed in the Integrated Forecasting System (IFS) as part of the European-funded SODA project (Hólm et al., 1999). Similar attempts were also promoted by other NWP centres during the same years. An ozone data assimilation was implemented in the NCEP Global Forecast System in June 1998 (C. Long, NOAA, personal communication). Struthers and colleagues (2002) discussed the first results from the assimilation of satellite-based ozone products in the UK Met Office Unified Model in their paper. An account of these early developments and results is available in Eskes (2003). These efforts demonstrated the feasibility of analysing AC fields exploiting an NWP framework; they spun off several studies to assess the added value of analysing ozone on the quality of meteorological fields and dynamics, and increased the confidence that, besides ozone, other constituents could have been considered as well. It was that increased confidence that led to the successful submission in 2002 of an ECMWF-led proposal to develop a comprehensive atmospheric composition monitoring and forecasting system (GEMS, Hollingsworth et al, 2008) followed by three European Union Framework Programs research projects (FP6-MACC, FP7-MACC-II, H2020-MACC-III). As part of GEMS and MACC, the IFS was extended to forecast and assimilate reactive trace gases such as tropospheric ozone (Flemming et al. 2015, Inness at al. 2015), CO2 and CH4 (Engelen et al. 2004, Agusti-Panareda et al., 2014), and aerosols (Morcrette et al. 2009, Benedetti et al. 2009). After a decade of developments, the AC component in the IFS had reached the level of maturity required to become the operational global model to monitor and forecast atmospheric composition as part of the ECMWF-led CAMS service funded through the European Union (EU) Copernicus programme in 2015.

The developments implemented within the GEMS and MACC projects, and now in CAMS, are fully integrated in the ECMWF IFS and are thus usable in NWP. However, the computational resources required to run the IFS with a fully integrated chemistry are too high to be exploited at the current NWP operational resolution (e.g. TCo1279 L137 for the high-resolution suite at the time of writing), and the level of complexity perhaps not necessary. For instance, the costs of running an IFS forecast would approximately increase by a factor of 4 if the tropospheric chemistry used in CAMS was adopted, and by a factor of 1.7 if all the aerosol species analysed in CAMS were to be considered. In contrast, negligible cost increase is anticipated if the most mature CAMS green-house gases, CO2 and CH4, were included. For this reason, the CAMS operational suite runs a global operational analysis and forecasts for reactive gases and aerosols at a low-resolution of TL511 L60 while a lagged suite for CO2 and methane is run at the split resolution of TCo399 L137 for the analysis and TCo1279 L137 for the forecasts. Therefore, affordable representations must be made in such a way that their realism is sufficient to lead to NWP improvements. To date, efforts have been devoted to construct ozone and aerosol climatologies from the corresponding prognostic variables generated by IFS using a recent version of the CAMS configuration. These climatologies are regularly updated to keep them consistent with the CAMS ozone and aerosols. Retaining such a consistency will be beneficial for a successful transition from climatology to the corresponding prognostic AC field in the radiation scheme.

It should also be recognized that added value could be independent from NWP scores, e.g. the availability of products of interest to users and Member States. For instance, high spatial resolution ultraviolet-index and surface visibility could benefit from prognostic ozone and aerosol. There is an increasing use of surface solar radiation forecasts by the solar-energy industry, which could potentially benefit from having prognostic aerosol (e.g. Schroedter-Homscheidt et al, 2013).

The present document suggests priorities for AC developments that can potentially benefit the NWP system. It summarises pan-ECMWF discussions based on the current knowledge and understanding, as well as experience acquired through research and development activities, including those that led to CAMS. AC is also relevant for C3S, namely to feed into reanalysis productions and to contribute to their seasonal forecasting suite. The lessons learned from the assessment of both the ERA5 reanalyses and the seasonal experiments run in preparation for the Seasonal System 5 (SEAS5) productions also provided important feedbacks on the current system and suggested possible areas where improvements might be achievable. These fed into the present document.

The following considerations are also noted:

1. the CAMS operational suite with a fully integrated chemistry represents today's upper limit of what can be affordable by and necessary in NWP, and thus that **the list of priority developments for NWP needs to be determined within the CAMS system complexity.** The CAMS system will serve as test bed to assess the added AC value on NWP for species not included in the current NWP configurations.

2. Atmospheric composition is the main concern for the CAMS. Thus, **AC developments that are important for the CAMS operational system may differ, especially in complexity, from AC developments that aim at improving the NWP system.** That said, CAMS is a user-driven service, thus any improvement of the NWP system based on the CAMS one and its products will be an important aspect to drive future CAMS developments.

3. New EU funding opportunities, e.g. on CO_2 monitoring (CHE project, www.che-project.eu), can pave the way to improvements in the IFS that could not be anticipated until recently (in that case on the IFS land component).

Based on these considerations, the discussions that led to the present document focussed on ozone (chapter 3), aerosols (chapter 4), and CO2 (chapter 5) to identify aspects for priority investigation and possible future implementation. The level of maturity and usage of these variables is substantially different from one to another, and the scope here is not necessarily to homogenise them.

Arguably, future scenarios and AC priorities may also depend on developments in other areas. For example, improvements in the infrastructure and numerical aspects could pave the way to developments that at the time of writing are too expensive to be contemplated in the operational set-up. These are discussed in Chapter 6. If an increased complexity of AC variables is implemented, then the Research-to-Operation process (R2O, Buizza et al, 2017, 2018) will need to be adapted accordingly, these considerations are presented in Chapter 7 for future reference. Finally, chapter 8 summarizes the recommendations and provides, from an AC perspective, a possible scenario for the IFS at the 2022 horizon.

2 Atmospheric Composition in a NWP system

Any model that aims at representing realistically the Earth's energy cycle and having a closure of the radiation budget needs to account for several atmospheric species.

For AC species that can have an impact on the NWP, the level of complexity required to represent them in an NWP system depends on the level of variability of the AC constituent itself, and on their dependence upon initial conditions. For slow varying species with no interannual/intraseasonal variability, a climatology will suffice. For those varying mainly at interannual time scales, a simple initialized prognostic capability, which could be as simple as damped persistence, is required. This may be the case of stratospheric volcanic aerosols. For those varying over a period of days and weeks a higher level of sophistication may be required. The latter should be modelled as prognostic variables interacting with physical processes (e.g. absorption/scattering of radiation), and thus with the main meteorological fields.

The accuracy of the prognostic species will depend on the quality of the initial conditions (ICs) and/or the representation of the emissions, the removal by wet and dry deposition, and chemical and physical conversion between species. For instance, the impact of some aerosols species (like dust or biomass burning) at monthly or long range may be more dependent on the emissions than on the ICs. These emissions may be either a result of the air-land/air-sea interactions, already included in the IFS, or may need to be modelled stochastically. In contrast, other species, like stratospheric ozone, may merely show a dependency on the ICs. The ICs will have to be provided for the real time and the reforecast. For the latter, these could be provided by AC reanalysis records. For the former, the IC quality can be improved via the assimilation of AC observations in the form of both level 1 radiances and level 2 retrievals.

Through data assimilation, the inter-dependence between AC and meteorological fields, particularly temperature and winds, can be reinforced and occur in different ways and through different mechanisms. The assimilation of radiances in spectral regions where the primary absorber is not a well-mixed gas (e.g. ozone) induces adjustments in both the primary absorber and temperature (e.g. McNally and Vesperini, 1996; Peubey and McNally, 2009). The presence of aerosols can contaminate the atmospheric signals and alias in temperature adjustments (e.g. Eresmaa et al, 2017). The application of the dynamical equations can induce changes in meteorological fields, particularly the winds. This is known as **tracer effect** (Peubey and McNally, 2009; Semane et al, 2009; Lupu and McNally, 2013). It is also worth mentioning that two other mechanisms, the **balance effect** and the **cycling effect**, can also produce adjustments from one variable to others. The former refers to the use of background error correlations and balance relationships in 3D-Var and 4D-Var multivariate data assimilation schemes. The latter refers to adjustments to meteorological variables induced by changes in one field that are propagated in time through cycling from one assimilation window to the next.

Figure **1** schematically shows that AC species can be represented with various degrees of complexity (and/or coupling). When needed, such degrees of complexity can be increased using a step-wise process. In Figure 1, **fully interactive** refers to a complete coupling of the relevant AC species with the meteorological fields through both physical processes and 4D-Var. In contrast, **partly interactive** refers to a coupling of the relevant AC species with the meteorological fields through either physical processes or 4D-Var. Except for the assimilation (dashed box) that is more relevant up to the medium range, the scheme can be applied to all forecast ranges. On extended ranges, data assimilation can still have a role through the choice of the ICs if these are derived from a reanalysis.

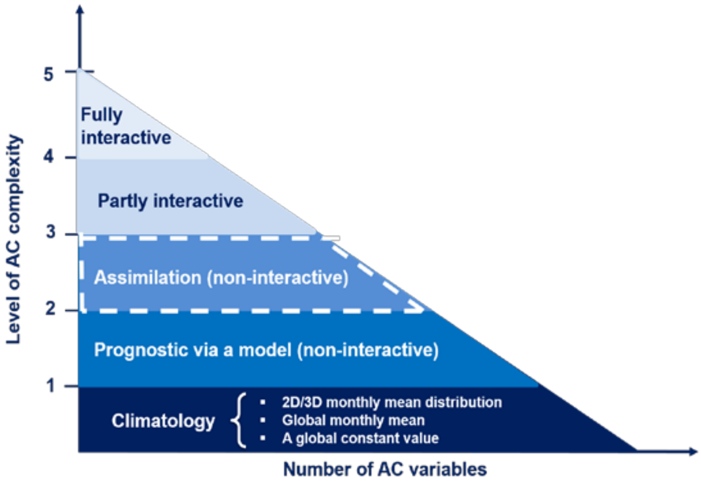

Figure 1: Schematic diagram showing that, when needed, an increase in the level of complexity is achieved via a step-wise process. The expressions "partly" and "fully" interactive refer to the fact that the mechanisms mentioned above, through which AC changes feedback on the meteorological fields, might or might not be all exploited.

3 Ozone

A fully interactive ozone (highest level of complexity in figure 1) can potentially add value to NWP applications. For instance, it can bring improvements in the stratospheric temperature (through the coupling with the radiation) and flow information (through the tracer effect) on the short to medium range, as well as a better representation of the QBO (e.g. Leblanc and McDermid (2001) used stratospheric lidar measurements to show the correlation between ozone and the QBO) and of the response to volcanic eruptions on longer forecast ranges (e.g. Ivy et al, 2017).

3.1 State-of-the-art in the IFS

Ozone is a fully integrated, prognostic variable in the IFS (Dethof and Hólm, 2004). In the NWP-like applications (medium-range weather forecasts, i.e. HRES and ENS, the ERA5 reanalysis, the ENS monthly extension and the seasonal ensemble SEAS), this prognostic ozone is used in the RTTOV calculations but neither interactively in their radiation scheme, which relies on a climatology from the CAMS reanalysis, nor in 4D-Var (through the tracer mechanism). Shortcomings in the ozone model have been identified (section 3.1.3) that need to be addressed before ozone can be successfully used interactively in the NWP-like applications. Data assimilation aspects, including bias correction and observation quality control, require careful reassessment with the currently available observations. In the CAMS operational suite, ozone (and aerosols) have been used interactively in the radiation scheme since June 2018.

3.1.1 Modelling ozone

In the NWP-like applications, ozone is modelled using a simple linear relaxation towards a climatological mean that is suitable for the stratosphere. In it, the ozone rate of change depends on the local ozone, the ozone column above the local point, and the local temperature. Two formally similar parametrizations are currently available, one described in Cariolle and Teyssèdre (2007) and referred to as the Cariolle scheme, and the other described in Monge-Sanz et al (2011) and referred to as the BMS scheme. There are two main differences between them:

1. the BMS relies on coefficients for the linear regression obtained from a 3D CTM as opposed to a 2D model used for those in the Cariolle scheme; and
2. The Cariolle scheme requires an explicit parametrization of the heterogeneous chemistry included as an additional term in the linear regression, not required in the BMS, as based on a CTM that includes a full description of the heterogeneous chemistry.

At the time of writing, the Cariolle scheme is used in the medium-range and monthly weather forecasting systems, and in ERA5; the BMS scheme is used in the seasonal forecasting system.

The IFS configuration of the operational CAMS system applies a tropospheric chemistry mechanism with 56 chemical species (Flemming et al. 2015), and the Cariolle scheme for the stratospheric ozone. Further options for the IFS in CAMS research configurations are different chemical mechanisms for both the troposphere and the stratosphere with up to 120 chemical species (a complete list of AC schemes available in IFS can be found in appendix C).

3.1.2 Ozone assimilation

In NWP, ozone is constrained via the assimilation of both ozone-sensitive radiances in the IR spectral range (IR/O3) and level 2 ozone products retrieved from UV measurements. Although technically possible, no level 2 ozone product retrieved from either microwave or IR measurements is currently assimilated in NWP. The assimilation of IR/O3 (Dragani and McNally, 2013) is performed together with all other radiances using the RTTOV radiative transfer model. The latter relies on the use of the prognostic ozone for its radiative calculations, and recent results showed that using a climatological ozone can degrade the system, e.g. in the fit to other used observations. The assimilation of ozone retrievals is performed assuming that the observation operator, which transforms the model state in an observation equivalent, can be approximated as a box-car function (in practice, for any given observed vertical layer, the used function is zero everywhere except over the layer it refers to, where it is equal to one).

There are no technical differences, at the time of writing, in the way ozone observations are assimilated across different IFS applications. However, differences may exist in the observations used (for instance HRES relies on the assimilation of IR/O3 not used in CAMS, and the latter assimilates MLS ozone profiles not used in the HRES), and in the way the background error statistics are computed. The latter are based on a climatology computed from an early ensemble of forecast differences for CAMS while they are directly provided by the EDA in the HRES in NWP. A better understanding than currently available is still needed to determine the ability of the EDA to derive the ozone background errors and to perturb the ozone field for the ensemble system. Some encouraging results have been obtained from a recent assessment of the value of doubling the number of EDA members from 25 to 50. This showed a more significant dependence of ozone background error estimates on the ensemble size than that of other meteorological fields. This resulted, for instance, in an improved level of agreement between the ozone first-guess / analyses and the assimilated ozone observations. Considering the encouraging result produced with the 50-member EDA, this aspect will be further investigated.

Quality control is applied to ozone data as done for all other observations. This includes initial checks on the observation metadata (e.g. on time and geolocation), a first-guess check to filter out data that are too different from the model background, and variational quality checks (VarQC) that are implemented during the analysis. While the ozone first-guess check has been tightened in recent years, the most recent developments in VarQC (e.g. application of the Huber norm; Tavolato and Isaksen, 2014) have not been explored for ozone. This could be an area for future improvements.

To correct for systematic biases, a variational bias correction (VarBC), like the one used for the radiance assimilation (Dee, 2005; Auligné et al, 2007), is used for ozone and anchored to the SBUV ozone profiles (Dragani, 2009). This strategy was tested and implemented in 2009, and may need to be revisited, especially as new ozone observations become available (for instance, those from the Sentinel missions) while existing instruments (like the SBUV) approach their end of life. This also requires that the observing system is kept up-to-date, and that ozone observations from new instruments are promptly included in the IFS. Although the latter is generally true, finding a replacement for the SBUV as VarBC anchor is urgent.

Work is on-going to improve the observation operator representation for level 2 products by means of the observation averaging kernels, which informs on the retrieval vertical resolution, and to test the feasibility of including a better characterization of the ozone observation uncertainties by accounting for the vertical correlations as provided by the data provider.

New generation instruments, particularly from the atmospheric Sentinel missions, will provide an unprecedent wealth of data at smaller than 10-kilometre horizontal resolutions. Data assimilation developments, for instance to account for spatial observation error correlations, are needed to fully exploit the information content of these high-resolution observations.

3.1.3 Impact of interactive ozone

Albeit all aspects of the ozone system are technically fully integrated in the IFS, ozone is not yet used interactive neither within the radiation scheme nor within the 4D-Var scheme (tracer effect from the retrieval assimilation) in the NWP-like applications.

The tracer mechanism, which could extract flow information from the assimilation of ozone products, has recently been assessed with the current observing system (modest in number of observations if compared with all the radiance data). Results show a generally neutral impact on the meteorological forecast skills. The wealth of data that will become available in the coming years (e.g. with the Sentinels satellite dedicated to the AC, and other missions like the TEMPO and GEMS geostationary satellites) has the potential to demonstrate the benefit of this mechanism.

At the time of writing, the radiation scheme (Hogan et al, 2017) in the NWP-like applications relies on a 2-D (latitude-pressure) monthly ozone derived from the CAMS interim reanalysis (Flemming et al, 2017). The impact of using an interactive ozone on the IFS meteorological fields has been evaluated in recent cycles using both the 4D-Var configuration, and the seasonal forecasting system.

When ozone is used interactively with the radiation in a 4D-Var configuration (Cariolle scheme, Cy42r1), the impact is neutral on the tropospheric forecast scores, and positive on the stratospheric scores, except near the Stratopause. This assessment was carried out with the old radiation scheme and needs to be reassessed in the current system.

In preparation for the seasonal ensemble SEAS5, which has been implemented in operation in November 2017 (Stockdale et al, 2018), experiments were run with both the Cariolle and BMS schemes, and for each scheme in both coupled and uncoupled configurations in the radiation scheme. Comparisons with ozone-sonde data showed that the BMS scheme produces better ozone forecast than that from the Cariolle scheme in lower stratosphere. However, the BMS scheme also gives insufficient total column ozone, and the tropospheric ozone is too low. The low tropospheric ozone is a possible reason for the apparent negative impact on scores of using the ozone radiatively interactive, although conclusive evidence on this is not available due to the very high cost of running sufficient size ensembles to investigate further. Based on these results, it was decided to replace the Cariolle scheme with the BMS in SEAS5, to obtain the best lower stratospheric ozone, which plays a role in delineating the tropopause definition used to set the vertical distribution of volcanic aerosol, but to retain the standard medium-range setting of not using the modelled ozone in the radiation. One consequence of not having an interactive ozone is a degraded QBO (temperature) representation in SEAS5 compared to that of SEAS4 (Figure 2).

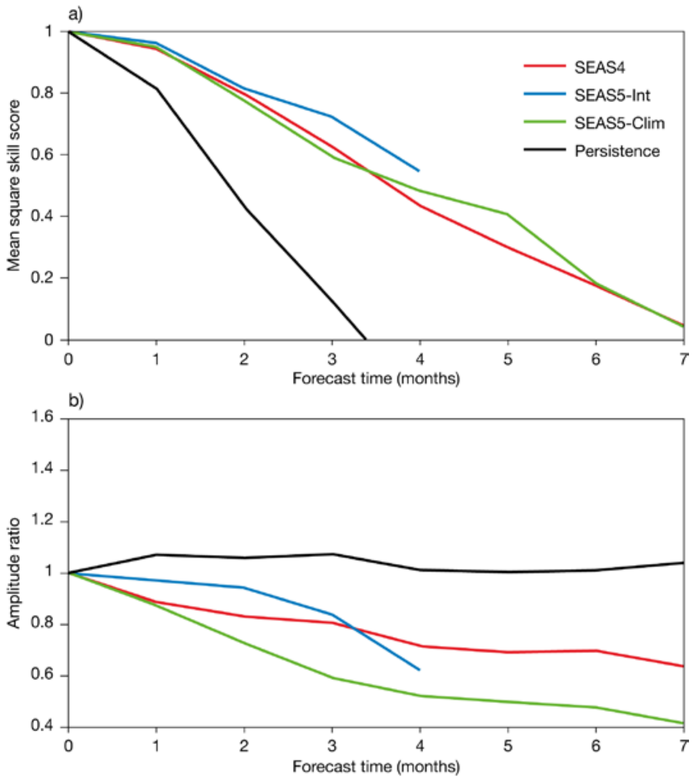

Figure 2: Statistics from forecasts of zonal mean temperature at 30hPa and between 5°S and 5°N, comparing SEAS4 (red) with prototypes of SEAS5 with climatological ozone (green) or radiatively interactive ozone (blue). Statistics include mean square skill score against climatology (top) and the amplitude of the signal relative to observations, which should be close to one (bottom).

3.2 Recommendations

It is recommended to move towards a fully interactive and seamless ozone.

To meet the above, the following aspects are noted and highly recommended:

- **Addressing the shortcomings of the ozone linear model at all forecast ranges is necessary** before ozone can be used interactively in the radiation scheme. Work to improve the ozone model ability in representing the mean state, and in the inferred tropospheric values is required. Detailed steps that can be undertaken are given in Appendix B.
- **Including new ozone observations, revising and improving the ozone data assimilation** (e.g. revising the VarBC and quality control, and assessing the adequateness of the EDA for ozone to

mention a few) **should also remain a priority** for medium-range applications. These improvements are necessary **to exploit the 4D-Var coupling through the tracer effect.**

Maintaining and improving the ozone climatology is also a necessity until prognostic ozone can be used interactively in the radiation scheme. Several improvements have been identified and are presented in Appendix A.

4 Aerosols

Aerosols have a key role in the atmosphere as through the absorption and scattering of the solar radiation they directly affect the Earth system's energy balance. The role that some aerosol species and the aerosol-cloud-radiation interaction could have in improving NWP systems have been increasingly tested in operational NWP systems. It is worth noting that the potential impact of the aerosol-cloud interactions (beyond the effect on cloud albedo through droplet number, i.e. first indirect effect) still needs to be quantified and their representation in GCM models may still be immature. The Met Office has been producing operational aerosol dust forecasts since 2008 and have a fully integrated dust in their operational data assimilation and forecasting system. Because mineral dust aerosol is the largest contributor to the global aerosol load and its variability occurs on temporal scales relevant for NWP, it is the species with the highest potential radiative impact in NWP, particularly close to the major dust source regions. Mulcahy et al (2014) showed that including interactive dust in the Met Office Unified Model (UM) improved model biases in outgoing long-wave radiation over West Africa. They also argued that including indirect radiative effects for all species but mineral dust and black carbon improved the surface radiation biases over part of Alaska due to lower cloud amounts in high-latitude clean-air regions with a positive impact on temperature and height. However, a computational cost-benefit assessment suggested the use of a climatology for the first indirect effect in the Met Office NWP system (Haywood, 2009). A review of the state of exploitation of aerosol for NWP forecasting is provided in Mulcahy et al (2014).

4.1 State-of-the-art in the IFS

In the NWP-like applications of the IFS, aerosols are not prognostic. Hogan et al (2017) discusses the representation and impact of aerosols in the IFS radiation scheme (direct effect). This relies on a 2-D (latitude-longitude) monthly climatology for aerosols developed from the CAMS interim reanalysis, and that replaces the Tegen et al (1997) climatology. Hereafter, this new climatology is referred to as the CAMS aerosol climatology. Bozzo et al (2017) discusses how the CAMS aerosol climatology was constructed and the NWP response following this update. A 3-D climatology including the vertical structure from the CAMS interim re-analysis is being considered for inclusion in 46r1. In extended forecast range simulations, aerosol information is also provided through the CAMS aerosol climatology (Bozzo et al, 2017). At the time of writing, all indirect effects are neglected.

A data assimilation and forecasting system for aerosols has been developed in the context of the GEMS and MACC projects (Morcrette et al, 2009; Benedetti et al, 2009), and further improved in CAMS. Such a development is fully integrated in IFS; thus, it offers an opportunity for NWP applications at different forecast ranges, and is described below.

4.1.1 *Modelling aerosols*

In the CAMS configuration of the IFS, aerosols are modelled using a bin scheme (Mocrette et al, 2009) that represents sea salt and desert dust (each sub-divided into three size bins), hydrophilic and hydrophobic organic matter, black carbon, and sulphate. With the current implementation, it is not possible to select and assess the impact of individual species out of those mentioned above. Work is currently on-going to increase on the one hand the aerosol model accuracy and on the other hand the level of flexibility for users to customize the choice of the available AC schemes, or to use only selected aerosol species as prognostic variables. Such a flexibility could pave the way to test e.g. the impact of selected aerosol(s) in NWP while providing the others through a climatology. A forthcoming development in the IFS is the implementation of the GLOMAP aerosol model developed at the University of Leeds (Spracklen, et al, 2005, Mann et al, 2010). This scheme predicts both number concentration and mass mixing ratio in several “modes” and, in turn, provides a description of the aerosol distribution. Thus, it is particularly apt to describe aerosol impacts on clouds as Cloud Condensation Nuclei can be predicted explicitly. For this reason, it was adopted in the UK Met Office climate model. Although its complexity makes it unaffordable at NWP resolutions, its availability could prompt studies to quantify the aerosol indirect effects, and possibly confirm recent results that a large volcanic emission of sulphate produced a measurable effect only on the cloud effective radius (first indirect effect, Malavelle et al, 2017), making this an aspect to consider as a future NWP development. These tests could also inform on a more physical parametrization of CCN than the current one (based on a surface wind-speed dependence using a different relationship over land and sea). The added value of such a parametrization could be compared with that obtained from using a climatology.

Also, major volcanic eruptions reaching the stratosphere can impact the stratospheric temperature distribution (e.g. due to absorption/scattering of radiation) for months after the eruption, thus they are important for seasonal prediction. With missing or incorrect vertical distribution of volcanic sulphate in the stratosphere, the stratospheric temperature response to major eruptions, like Pinatubo, is also incorrect (Figure 3).

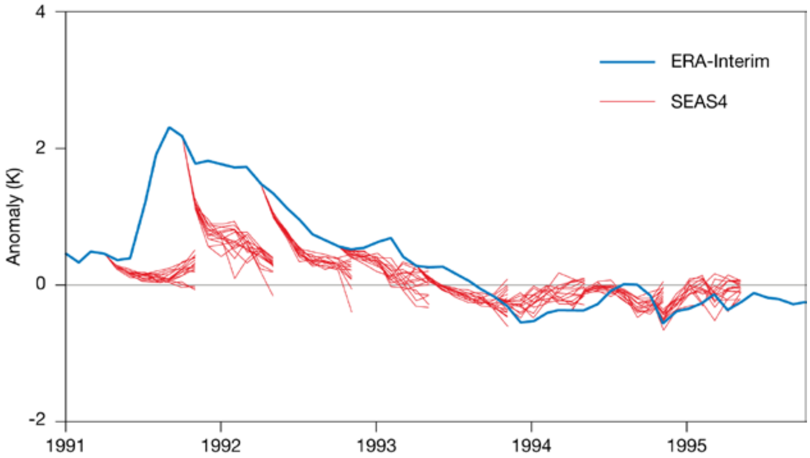

Figure 3: Temperature anomalies at 30hPa and at month 7 (1 May and 1 November start dates) following Pinatubo from SEAS4 (based on a 15-member ensemble).

4.1.2 *Aerosol assimilation*

The aerosol analysis, which is available under the CAMS configuration, is an extension of the 4D-Var analysis including the total aerosol mixing ratio as aerosol control variable, computed as sum of all contributing aerosol species. The aerosol background error statistics are based on a climatology calculated according to the Derber and Parrish (1992) method (Benedetti and Fisher, 2008) and recently updated on the 137 model levels, and thus they are not based on the EDA. The latter will need to be investigated should some aerosol species be included in the NWP operational suite. Increments to the total aerosol mixing ratio are distributed to the individual species according to their fractional contribution, which is maintained constant in the 4D-Var minimization. As the assimilation of aerosols is a severely under-constrained problem, contributions that should be attributed to missing species are aliased onto modelled species. Also, the speciation of the resulting aerosol analysis can differ considerably from the speciation of the modelled aerosol because of the different atmospheric lifetimes of the aerosol. For example, the relative contribution of absorbing black carbon aerosol can be exaggerated in the aerosol analysis. This aspect needs thorough investigation during the testing of the impact of prognostic and climatological aerosols on NWP. The aerosol vertical distribution is not constrained by the currently used observing system. An experimental development using two control variables, a fine and a coarse mode mixing ratios, is also available. Although never been used operationally, it is now being revived to benefit from assimilation of multi-wavelength aerosol reflectance in the visible.

The capability of assimilating aerosols is currently only exploited in the CAMS application. This routinely assimilates Aerosol Optical Depth (AOD) at 550nm from the MODIS instruments, and from combined MetOp-A and -B measurements produced by EUMETSAT. Assimilation of other AOD products was also tested. However, comparisons with independent data showed that they under-performed compared to MODIS, which remains the dataset with the highest impact.

Work is on-going to support the data assimilation system readiness and trial of new observation types, for instance lidar backscatter profiles from satellites and ground-based network. Lidar data could be important to constrain the aerosol vertical distribution, unconstrained by the currently assimilated observations. Worth-mentioning is that the joint assimilation of profiling instruments (like lidars) and imagers (e.g. MODIS) is still a scientific challenge due to their relative biases. The currently used bias correction scheme (based on VarBC) is likely too simple to account for these biases, and will require substantial improvements.

Quality control checks are applied to aerosol observations. The initial quality checks performed during the pre-screening and that are dataset-dependent are up-to-date for the currently used observations. The first-guess and VarQC, which are currently implemented like for most observations, could be revised using the latest developments should some aerosol species become part of the NWP operational suite.

With a high likelihood of a major volcanic eruption like that of Mt. Pinatubo in 1991, the readiness of the IFS data assimilation system is a major concern. Currently, the RTTOV radiative transfer model used in the IFS filters out all aerosol-contaminated radiances from the IR sounders. Therefore, it can be expected that, in case of a major eruption, a large amount of radiances from the region interested by the volcanic plume would be contaminated by aerosols, and hence discarded. A consequence of this could be that the medium-range forecasts, especially the tropospheric temperature, might be less constrained by the IR measurements, and potentially degraded. Work is currently on-going to support the development and testing of a demonstration data assimilation system of volcanic ash observations from in-situ lidars. While this work exploits the CAMS configuration of the IFS, it could be extended to the

NWP configuration. Enhancing the IFS system readiness in case of a major volcanic eruption at all forecast ranges would be advisable.

4.1.3 Impact of interactive aerosols

Tests using the prognostic aerosols in the radiation scheme were performed in the CAMS configuration, together with the interactive ozone (see section 3). Results showed degradation in the NWP scores while preserving the quality of the AC forecasts. As the primary products of the CAMS system are AC fields, it was decided to use the prognostic aerosol and ozone in the production of the CAMS re-analysis (Inness et al., in prep) and in the CAMS NRT o-suite from Cy45r1 (June 2018) onwards.

The impact of fully prognostic aerosols (all species, direct effect only) was also tested in a series of (Cy41r1 and Cy43r1) extended forecast range experiments for the May-October months from 2003 to 2015 (Benedetti and Vitart, 2018). Results suggest that radiatively-interactive aerosols, particularly desert dust and biomass burning, are modulated by the Madden-Julian Oscillation and may significantly increase model skill at week 4 compared to the simulations using either the Tegen et al (1997) or the Bozzo et al (2017) climatology based on the CAMS interim re-analysis. Furthermore, the analysis of specific events showed that, for instance, the 2015 6-month re-forecast (initialized on 1 May) exhibits high correlations between the large biomass burning patterns of radiative cooling at the surface (right panel in Figure **4**) and the anomaly linked to the fires during the anomalous Indonesian fire season (left panel in Figure **4**). In these simulations, observed fire emissions were used while dust emissions were calculated from model winds. This would suggest that improved 2m temperature seasonal forecasts could be obtained if radiatively-interactive aerosols were coupled with a prognostic fire emission model.

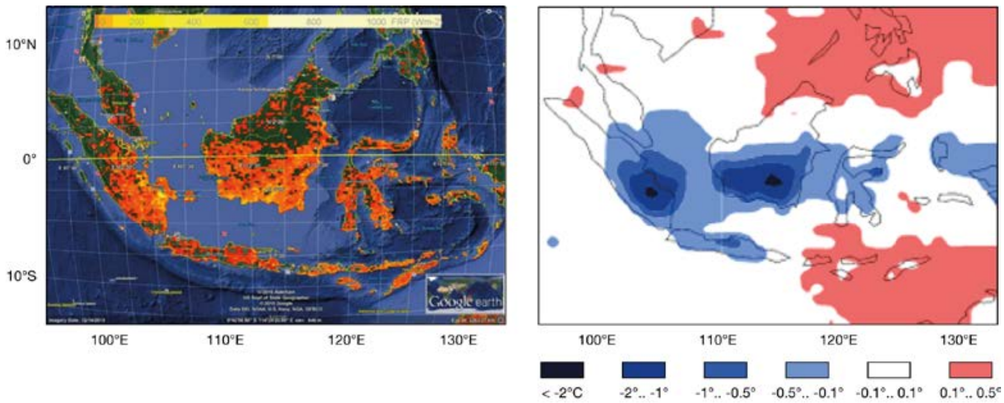

Figure 4: Fire radiative power for the Indonesian area for Aug-Oct 2015 (left) and 2m temperature anomaly showing the radiative cooling induced by the smoke aerosols which was well captured by the interactive aerosol run for October 2015 (right). This change in 2m temperature could not have been captured without the prescribed fire emissions.

4.2 Recommendations

There are indications that including some aerosol species interactively in the IFS could be beneficial in NWP-like applications. Detailed assessment of the added value of individual species is not yet possible. On-going work towards an increased flexibility of the IFS in the CAMS configuration is a major technical stepping stone to facilitate impact assessment of the direct and first indirect effects of selected aerosol species.

The following recommendations are made

- **Perform a detailed assessment of the added value of using selected prognostic aerosol species (direct effect) interactively at all timescales**.
- **Test the benefits of using a climatology for computing the effective radius (1st indirect effect).**
- **Investigate the data assimilation of aerosol information (retrievals and radiances) of selected species** (e.g. dust) if results from the above tests indicate that for those selected species a higher level of complexity/coupling than a climatology is beneficial for NWP applications.
- **Improve the representation of the decadal variations of prescribed aerosol data sets and the aerosol climatology** (specific recommendations for the latter are presented in Appendix A). These efforts should, when possible, build on CAMS reanalysis. This activity will need to continue even if some prognostic aerosol species were selected to be used interactively.

5 Carbon Dioxide

Representing time-varying carbon dioxide is important for capturing climate trends, and to correctly represent variations in seasonal prediction and decadal/multi-decadal climate prediction. Arguably, the CO_2 impact in global NWP models is less well understood. Engelen and Bauer (2011) presented an assessment of the impact of CO_2 in NWP. Their results, based on an old version of the IFS that did not account for the surface variability, showed that having a variable CO_2 reduced the first-guess biases of the IR temperature-sensitive radiances. Sizeable improvements in the CO_2 modelling and assimilation capability allows to revisit those studies considering the realism gained by CO_2 in simulating atmospheric variability (Agustí-Panareda et al, 2014, 2016, 2017), as illustrated in Figure **5**.

The attractiveness of better representing CO_2 in the IFS is twofold. On the one hand, it could help reducing the observation bias associated with the assimilation of temperature soundings in the longwave, where CO_2 is the major absorber, and, on the other hand, it could improve the Earth System Model realism of the IFS by representing surface emissions in its land component and their interaction with the meteorological fields. For example, the inclusion of the interaction between the carbon and water/energy cycles would allow the use of observations of photosynthesis (e.g. Solar Induced Fluorescence from a range of satellites) to improve latent/sensible heat fluxes and CO_2 uptake by vegetation concurrently.

Worth-noting is that representing CO_2 does not require any complex chemistry development. Therefore, it is an ideal tracer that could also serve to verify the ability of IFS to conserve mass (Agustí-Panareda et al. 2017, Diamantakis and Agusti-Panareda, 2017) and to develop Observing System Simulated Experiments for data assimilation approaches that simultaneously correct concentrations and surface fluxes.

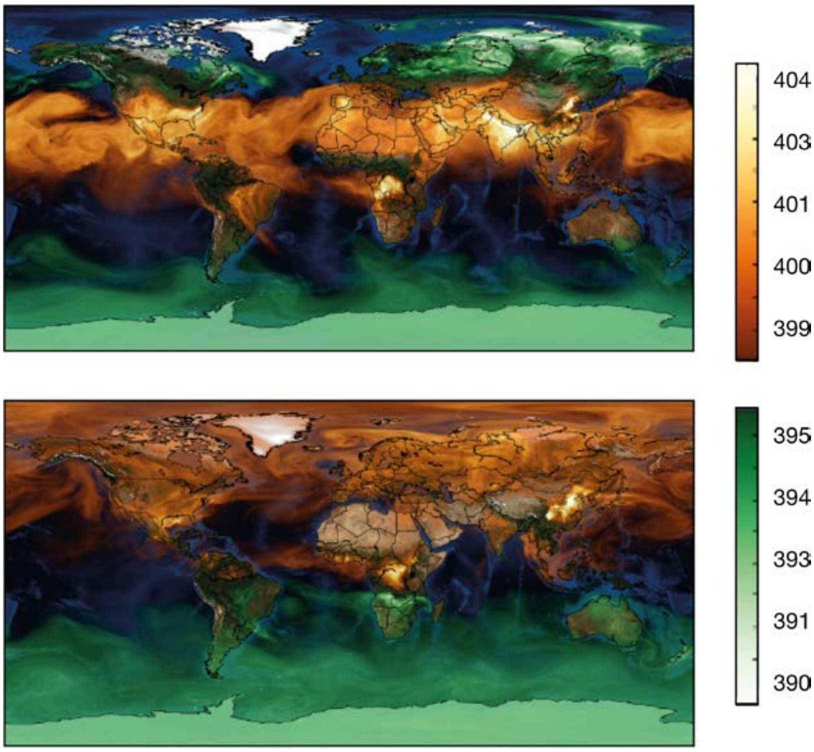

Figure 5: CO_2 [ppm] concentrations on 15 January (top) 2015 and 15 July (bottom) at 00 UTC showing values above and below the global mean in reds and greens respectively (see colour bar), obtained with the Cy43r1 HRES 9km 1-year nature runs (CHE project).

5.1 State-of-the-art in the IFS

Carbon dioxide is hardly exploited in the NWP-like applications of the IFS. However, CO_2 can affect aspects of the IFS. As mentioned above, a good representation of this species in RTTOV can be beneficial for the assimilation of the temperature soundings in the longwave spectral region. The RTTOV model currently relies on an out-of-date global mean CO_2 profile that does not reflect current CO_2 levels. Ideally, not only would the CO_2 information used to determine the model equivalent of the radiances in RTTOV reflect current CO_2 levels, it would also reflect the geo-variability of its sources and sinks. Figure 6 shows an off-line calculation of IASI first-guess departures (O-B) obtained with two CO_2 representations in RTTOV: the global mean profile used in HRES (black) and one obtained by scaling it with an observation-based climatology (red). Improving the CO_2 representation leads to a reduction of a few tenths of a degree in the observation bias, confirming the results from Engelen and Bauer (2011).

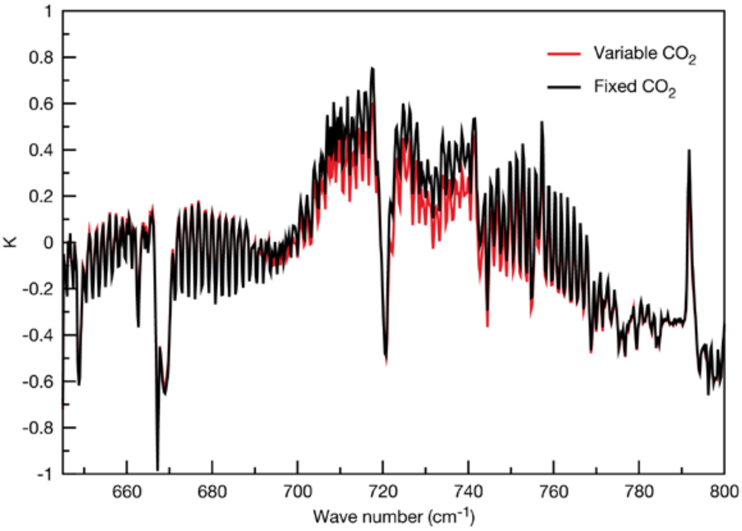

Figure 6: Mean first-guess departure (O-B) for IASI measurements with the first-guess equivalent of the observations computed using two CO_2 representations in RTTOV. The black line refers to the current HRES conditions; the red line shows the O-B departures when the CO_2 profile is scaled according to a CO_2 climatology obtained from the ground-based measurements.

5.2 CO_2 emissions and surface processes

The surface emissions (or fluxes) are a key element for the spatial and temporal concentrations in atmosphere. Emissions that show diurnal cycle variations are normally treated as prognostic variables connected to energy and water variables as predictors.

The natural ecosystem exchange of CO_2 can be considered prognostic as surface flux interacts with the meteorological forcing (Boussetta et al, 2013). However, large uncertainties characterize the variability of both natural and anthropogenic CO_2 (IPCC, 2013), especially at the surface.

Realizing a coupled assimilation of CO_2 including capability of correcting surface emissions is one of the scopes of the CHE project.

5.3 CO_2 modelling and assimilation

A global atmospheric CO_2 data assimilation (Engelen et al. 2004, 2009) and forecasting (Agustí-Panareda et al, 2014, 2016, 2017) system was implemented in IFS as part of the GEMS and MACC projects, and now further improved in CAMS. Massart et al (2016) extended the initial data assimilation system to exploit GOSAT retrievals. It is noted that, at the time of writing, satellite retrievals of CO_2 are available with a delay of about 3 days. Forthcoming CO_2 datasets, for instance from the OCO-2 mission, could be provided in NRT.

One of the strengths of the CO_2 forecasting system is that the land surface, including vegetation CO_2 fluxes, is modelled online within the IFS using the CHTESSEL/CTESSEL land surface scheme (Boussetta et al, 2013). Other CO_2 fluxes are prescribed from inventories and from off-line statistical and physical models. The CO_2 anthropogenic emissions are from the EDGAR inventory (http://edgar.jrc.ec.europa.eu). This system constitutes the basis of a NWP-system that incorporates CO_2 monitoring capacity as illustrated in Figure **5**. The CO_2 forecast also benefits from the transport modelling from a state-of-the-art numerical weather prediction (NWP) system initialized daily with a wealth of meteorological observations. This system constitutes a first prototype of a NWP-system that incorporates CO_2 monitoring capacity.

A newer release of the CO_2 inventories (EDGAR4.3.2, Janssens-Maenhout et al. 2017) and a new characterisation of surface emission uncertainties are the base of CO_2 coupled data assimilation system, which is being developed using the OOPS (Object-Oriented Prediction System) framework.

5.4 Recommendations

Important developments to the CO_2 data assimilation and forecasting system are expected in the next few years, driven by CAMS and the CHE project.

With the current state of play and until such developments are implemented **it is highly recommended to improve the CO_2 representation in RTTOV** using a more realistic global CO_2 profile while a CO_2 climatology from the CAMS reanalysis can be developed.

With a lower priority than the above recommendation, it is noted that it could be beneficial:

- **Running an explicit CO_2 tracer in the NWP suite to assess the IFS transport patterns**, provided the robustness of the CO_2 system. The associated computational cost on the NWP operational system would be small allowing to integrate a key essential climate variable in the IFS, with the potential to couple to RTTOV in the NWP analysis and exploit interaction with radiation in the forecast.
- **Revising the biosphere CO_2 flux representation within CHTESSEL/CTESSEL** for improving the surface latent/sensible heat fluxes and their partitioning.

6 Infrastructure, code efficiency and dynamical core developments

There are several numerical developments that could be exploited if an increased complexity of AC representation in NWP-applications was considered to offset, at least in part, additional computational costs that could be generated. These are discussed below.

6.1 Maintenance and standardization of the AC code in IFS

The developments performed through an ever-increasing number of applications and projects over the last 1-2 decades on one hand have increased the capability of the IFS and the availability of new fields, on the other hand have also increased the numerical complexity of the source code. This is particularly the case of AC. For instance, ozone can currently be analysed through two different paths, one used in the HRES and the other used in CAMS leading to some code duplication, e.g. each application refers to its own ozone observation operator (that transforms the model field into an observation equivalent in 4D-Var) even though the latter is essentially the same.

This is because all AC developments for CAMS are under dedicated switches. Such a choice was motivated in the past by the ECMWF's need to develop the chemical components in preparation for the CAMS service while maintaining the NWP code bit identical. Today that both are operational services, this approach can be rethought. Although restructuring part of the AC code might not necessarily save computational costs, it would improve the efficiency in code maintenance and code improvements.

Furthermore, if AC variables are to become a bigger part of the NWP operational system than they are currently, then additional aspects in the AC infrastructure may require attention and standardization. These include all aspects of observation data handling (data pre-processing, initial quality control and thinning that need to be performed within the COPE environment as for all other observations), variational bias correction, data assimilation developed as in the OOPS framework, and EDA-based background error covariances. While some of these aspects (e.g. data handling using the COPE environment) are only required for the operational implementation, others can affect the scientific results of the testing, and therefore will need to be developed before the latter can be performed (e.g. bias corrections). Among the latter, there might be developments that while affecting the scientific results could be replaced by cheaper and ad-hoc solutions for the initial testing. For instance, one could assess the impact of individual aerosol species using them static background error covariance matrices, and develop far more complex EDA-based ones only for the selected aerosol species at a later stage.

6.2 The multiple grid development

The introduction of the Atlas library by Deconinck et al (2017) has paved the way to novel numerical method developments for the IFS. One development relevant to AC forecasts is the option to run certain IFS processes using a dual grid. The aim is to offset the cost of an expensive model process by simulating it on a grid with reduced resolution. This technique has been successfully used for over a decade for the radiation scheme and, together with the Atlas library, recently applied to the IFS tracer transport scheme (it is expected to be released in cycle 46r1). The dual grid formulation allows tracer advection on a separate lower resolution grid using either a computationally cheap semi-Lagrangian option or the Multidimensional Positive Definite Advection Transport Algorithm (MPDATA, Kühnlein and Smolarkiewicz, 2017). The latter is a finite volume mass (FVM) conserving transport scheme used by the new dynamical core (Smolarkiewicz et al, 2016) developed at ECMWF.

The MPDATA time-step is computationally more expensive than the unconditionally stable semi-Lagrangian (non-conserving) scheme. However, by reducing the resolution of the grid on which the advection takes place, a significant portion of this extra cost could be offset. Additional cost savings can be obtained by taking advantage of code optimizations recently tested as part of the scalability programme and by improving the efficiency of the vertical advection scheme (e.g. implementing the dimensionally-split advection scheme which has been recently developed in FVM dynamical core and allows doubling of the advection time-step).

Improvements in the MPDATA accuracy are already envisaged, e.g. by reformulating its equations from a time-dependent height coordinate system to the fixed “eta” coordinate system used by the semi-Lagrangian advection scheme of the IFS, and by improving the accuracy and conservation of the horizontal interpolation scheme (used to map the tracer fields from the low to the high-resolution grid and vice-versa). With these developments in place, it would be possible to perform a scientific evaluation and examine the benefits of the mass conserving flux-form finite-volume approach against the current semi-Lagrangian approach under a unified model framework. This will be a unique strength of the future IFS.

If, on the one hand, the potential cost saving achievable with a multiple grid approach is attractive, on the other hand, it may not be as straightforward for AC as it was e.g. for the radiation scheme. The AC source and sink modelling developments are not self-contained in a module coupled with the physics at one time, but rather intermingled with it. For instance, the injection of the emissions is parametrized in the diffusion scheme; the convective transport is part of the convection scheme; loss processes, such as wet deposition, need inputs from the precipitation physics; and the dry deposition and biogenic CO_2 parametrization rely on input from the land surface scheme. Worth-noting is that using different grids to simulate the tracer transport and the chemistry and aerosol source and sink terms could also mean to have the tracer not collocated with its tendency (e.g. Flemming et al, 2009). Thus, for the multiple grid solution being feasible the AC sink and source calculations should be performed on the coarse grid.

6.3 The single precision development

A single-precision (SP) version of the IFS (Váňa et al, 2017) will become operational on the new supercomputer. Tests show that it can reduce the computational cost of forecast jobs by a factor of 40%. Although, this development was not thought for AC, it could be considered as a cost-saving possibility if more complex AC configurations than currently used were adopted in NWP.

Recent tests with AC forecasts with the tropospheric chemistry scheme used in the operational CAMS configuration has shown that SP chemistry code produces results that are not neutral compared with its double precision (DP) equivalent. Thus, work is required to refine the code and define a set up that exploit the SP cost-saving option for AC forecasts while maintaining the quality of their DP equivalents as in the case of weather forecasts.

6.4 The semi-Lagrangian departure point (SLDP) calculation

Improvements in the SLDP calculation scheme and the interpolation methods are continuously investigated with potential to improve cross-tropospheric transport and reduce biases introduced by the operational scheme. Any improvements in this area will automatically benefit AC forecasts.

Worth noting is that SLDP is accurate and computationally efficient in a single grid configuration. The costs of the SLDP do not change by increasing the number of tracers, as the departure point and the interpolation weights are only computed once. However, the cubic interpolation must be applied as many times as the number of tracers considered. A drawback of the SLDP method is that it is mass conservation deficient. The implementation of a mass conserving advection scheme that improves the current standard is highly relevant and of interests for all IFS applications.

6.5 IFS code optimization and configuration

A detailed cost analysis of the AC overhead computational costs is still needed, though a preliminary assessment shows that the relative costs significantly depend on IFS configuration and resolution considered. For instance, in the IFS monthly forecast runs coupled with the NEMO ocean model, the overhead of the 12-bin aerosol scheme is about 10% (factor 1.1). If this exercise was performed with an uncoupled ocean model the increase in costs due to the aerosols would be 70% (factor 1.7). The overhead of running the uncoupled IFS with AC in data assimilation is lower (factor 2) than the overhead for a forecast run (factor 4).

Despite these differences, a preliminary assessment from a forecast run seems to indicate that only between 50-70% of the additional costs are due to the AC source and sink calculations. The remaining overhead is required for I/O operations and to run AC diagnostics. Thus, some cost savings could be immediately obtained if some of these diagnostics were switched off and by improving the I/O efficiency.

6.6 Recommendations

The following work is highly recommended:

- **Harmonize the duplicated ozone fields in the IFS.**
- **Refine the AC-related IFS code and define a set up that exploits SP for AC** while maintaining the quality of DP fields as in the case of weather forecasts.
- **Perform a detailed cost analysis of the actual overhead caused by an increased complexity of AC, particularly aerosols.**
- **Improve the implementation of the mass conserving MPDATA advection scheme** included in the IFS multiple grid development, **assess its impact and cost for AC forecasts.**

The work to further develop and test the multiple resolution-grids as an option for cost-efficient AC simulation is regarded as a medium priority goal for the forthcoming four years.

7 Diagnostic tools and R2O process

If AC variables are to become a bigger part of the operational system than they are currently, they need to undergo the same range of verification and diagnostics as the main thermodynamic and wind fields. Thus, verification and diagnostic tools that are routinely used in the R2O process (Buizza et al, 2017, 2018) should be extended as appropriate and provide relevant AC fields as a default to provide:

- **Analysis-based reference:** verification of the operational HRES forecast against either its own analysis or the operational CAMS AC fields (provided possible issues related to the different spatial resolutions can be overcome). It has the advantage of incorporating a forecast model and all available observations to provide the best estimate of the state, globally, albeit lacking independence from the system being verified. For AC, it has the disadvantage of relying on AC analyses that are less constrained by observations than meteorological fields.
- **Observation-based reference:** verification of the first-guess forecasts against observations that are assimilated (or passively monitored) within an analysis experiment (online) and against reference observations (offline). The advantage of **observation-based verification** is its independence when the verifying observations are not assimilated. Various offline tools have been developed for CAMS and available for NWP purposes. The disadvantage is that reference observations are often provided by in-situ network (thus they cannot provide a global and homogeneous coverage), and, in some cases, with important time delays (e.g. most ozone sondes are made available only months after the launch).

Additionally, **process-based diagnostics**, such as monitoring the effect on the QBO in seasonal forecasting, could also be contemplated.

7.1 Recommendations

The following actions are noted and recommended for future discussion and possibly implementation in the R2O process if the level of complexity/coupling of AC species should increase:

- **Archive NWP-relevant AC fields routinely in operations and research experiments** and **include model-data pre-processing and archiving tasks required by the** available **diagnostic tools** in the RD IFS suites. This will have a cost due to the additional MARS archiving capability required and will need to be quantified.
- **Extend tools routinely used for verification to NWP-relevant AC constituents** as standard; **use CAMS operational analyses as the reference**. The latter may require adjustments, for instance to account for the different resolutions.
- **Catalogue, and assess the robustness, quality of documentation, and level of maintenance and user support of other AC-related diagnostic tools** (e.g. developed as part of the CAMS activities).

8 Summary: priority developments for a 2022 implementation

Atmospheric composition is an integral part of the Earth system, affecting the radiative balance, hence the temperature, wind and mass distribution. Thus, its inclusion in a NWP system, like the IFS, can have potential impact on its predictive skills. The IFS includes detailed representation of several reactive gases, green-house gases, and aerosols that supports the CAMS service on atmospheric composition. However, level of complexity and corresponding computational costs are too high to run such a system at the NWP resolution, and thus to fully exploit the advantage of the available AC implementation. Targeted research is needed to identify the level of complexity and/or coupling for AC that can improve the NWP systems with solutions that are also computationally affordable.

An assessment of the system capability currently used by the CAMS application (used as upper limit) and of the state-of-the-art of AC development in NWP-like applications shows that steps can be taken to improve the latter and benefit from some of the existing schemes available within the former.

These will prepare for the IFS system used in NWP-like applications in 2022. The CAMS configuration can serve as an initial test-bed for species not included in the NWP configurations.

The following recommendations have been identified for ozone, aerosols and CO_2:

1. **Move towards a seamless fully interactive prognostic ozone**; fully interactive refers to the coupling in both the radiation scheme and in the data assimilation**.** This will require:
    a) **Addressing the shortcomings of the ozone linear model at all forecast ranges is necessary** before ozone can be used interactively in the radiation scheme (see Appendix B).
    b) **Including new ozone observations, revising and improving all aspects of the ozone data assimilation** (e.g. revising the VarBC and quality control, and assessing the adequateness of the EDA for ozone to mention a few) **should also remain a priority** for

short and medium-range applications. These improvements are necessary **to exploit the 4D-Var coupling through the tracer effect.**

Maintaining and improving the ozone climatology is also a necessity **until prognostic ozone can be used interactively in the radiation scheme** (see Appendix A).

2. **Improve the aerosol climatologies** (for direct effect and possible improved representation of first indirect effects) **and, through systematic testing, to seek demonstrating the benefit (at an affordable cost) of including a limited number of prognostic species** (e.g. dust). In detail, this means to:

   a. **Perform a detailed assessment of the added value of using selected prognostic aerosol species (direct effect) interactively at all timescales**.

   b. **Test the benefits of using a climatology for computing the effective radius (1st indirect effect).**

   c. **Investigate the data assimilation of AOD information of selected species** (e.g. dust) **and direct radiance assimilation** if results from the above tests indicate that for those selected species a higher level of complexity than a climatology is beneficial for NWP applications.

   d. **Improve the representation of the decadal variations of prescribed aerosol data sets and the aerosol climatology** (see Appendix A for the latter) building, when possible, on CAMS reanalysis. This activity will need to continue even if some prognostic aerosol species were selected to be used interactively.

3. **Enhance the CO_2 representation in the RTTOV** to improve the radiance assimilation.

Because any increase in complexity and/or coupling will most likely increase the computational costs of running the IFS, work on improving the efficiency and off-setting any additional computational costs will need to be pursued. To increase efficiency and offset AC-related extra computational costs, it is also recommended to

- **Harmonize the duplicated ozone analyses in the IFS.**
- **Refine the AC-related IFS code and define a set up that exploits SP for AC** while maintaining the quality of DP fields as in the case of weather forecasts.
- **Perform a detailed cost analysis of the actual overhead caused by an increased complexity/coupling of AC, particularly aerosols.**

It is also recommended to **design and implement a mass conserving advection scheme.**

The following actions are noted and recommended for future discussion and possibly implementation to facilitate the R2O process should the level of complexity/coupling of AC species increase:

- **Archive NWP-relevant AC fields routinely in operations and research experiments** and **include model-data pre-processing and archiving tasks required by the** available **diagnostic tools** in the RD IFS suites.

- **Extend tools routinely used for verification to NWP-relevant AC constituents** as standard; **use CAMS operational analyses as the reference**.
- **Catalogue, and assess the robustness, quality of documentation, and level of maintenance and user support of other AC-related diagnostic tools** (e.g. developed as part of the CAMS activities).

Without discussing it in detail, the present document recognizes that improvements might not only come as increased forecast skills. For instance, some of the recommendations given above could improve existing diagnostics and by-products of the IFS, like high spatial resolution UV and visibility, that could become of interests to some of the ECMWF stakeholders and perhaps be included in the ECMWF portfolio.

Additional aspects would also be beneficial. However, based on the current and foreseen level of resources over the next four years, they are considered as best-effort activities. These include:

- Work to further develop and test the multiple resolution-grids as a long-plan option for cost-efficient AC simulation;
- Use AC tracers in the NWP configurations for diagnostic purposes. For instance, radon could be used to diagnose transport and mixing processes, hydrogen cyanide (HCN) is emitted by biomass burning, which can lead to anomalies in the IR radiance monitoring; CO_2 can help verifying the IFS ability to conserve mass. The computational cost of a tracer is negligible;
- Enhance the IFS system readiness in case of a major volcanic eruption at all forecast ranges.

Work to further improve the IFS Earth System Model realism in representing surface emissions and their interaction with the meteorological fields could spin-off from developments included as part of the CAMS and CHE activities. Thus, any plan on this aspect will need to be revisited once the CHE project will have reached its conclusion and a clearer understanding than currently available of its achievements and follow-on project opportunities are known.

Finally, it is noted that the implementation of the given recommendations will rely on work of a relatively large group of experts across three departments (Research, Copernicus, and Forecast Departments) that will support it at their best effort. Although new funding opportunities aligned with the above recommendations will be sought, they might not become available within the forthcoming four-year period. Thus, an increased level of coordination of all AC-related activities that will better exploit scientific and infrastructure synergies, avoid duplication of work, and help planning developments of common interests will be beneficial to both the NWP and the CAMS applications.

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Appendix A: Improving AC climatologies for the IFS

Climatologies are a robust way to provide AC input to any parametrization, particularly the radiation scheme. Because the AC climatologies used in the IFS are mostly based on CAMS interim re-analysis, which is also produced with the IFS, they are more consistent with the prognostic AC fields than external climatologies. This consistency is an advantage because it could facilitate and speed up the R2O process in case the transition from climatological to prognostic representations is considered as less retuning is needed.

The production and assessment of the CAMS ozone and aerosol climatologies in the radiation scheme has indicated several shortcomings. The following list presents the known issues and possible improvements to be considered in future climatologies:

Mesospheric ozone: The current ozone climatology is based on the CAMS interim re-analysis from surface up to 0.1 hPa (CAMS model top), and extrapolated values in the mesosphere (0.1-0.01 hPa). Improvements of the climatology in the mesosphere can lead to improved representation of the ozone diurnal cycle, which shows about 30% lower values during the day than that during the night. Hogan et al (2017) showed that reducing day-time ozone in the lower mesosphere leads to reduced temperature biases.

Tropospheric ozone: In contrast to the mean stratospheric ozone that only shows latitudinal variations, the mean tropospheric ozone exhibits a significant spatial variability depending on the location of the emissions and global transport patterns (Ziemke et al. 2011). Thus, including a latitude-longitude dependency of the tropospheric ozone climatology, like for aerosols, will probably be beneficial.

Aerosols: In its original formulation, the CAMS aerosol climatology produced degraded temperature forecasts in the West Africa regions, where the black carbon burden is high. Producing an updated aerosol climatology in line with the latest updates to the CAMS system (e.g. the latest CAMS re-analysis, Inness at al, in prep) together with a consistent update of the aerosol optical properties is advisable.

The following aspects are also noted:

1. Representations of trace gases and aerosol are also required for the radiative transfer calculations used in the radiance assimilation code (RTTOV). An inventory and harmonisation of common data sets used in the model physics and data assimilation might be advisable.

2. Seasonal prediction and re-forecasts cover longer periods (1981 to present day) than CAMS reanalyses (2003-onwards). The underlying AC climatologies used in the former must account for trends (e.g. related to anthropogenic emissions) and inter-annual variability (e.g. due to major volcanic eruptions). The time-varying AC forcings currently used in ERA5 and seasonal forecasts are the recommended CMIP5 data sets. Ensuring consistency between the CMIP data and the aerosol and ozone climatologies used in the NWP configuration would be beneficial for a seamless prediction.

Appendix B: Improving the linear ozone scheme

The linear ozone scheme of the IFS is primarily appropriate for representing stratospheric ozone chemistry, and not suitable for the troposphere. This limits its usefulness in the radiation scheme because, for instance, the longwave absorption by tropospheric ozone in the Tropics cannot be adequately modelled. As discussed in section 3.1, two formally similar linear schemes are available, each with its own shortcomings.

To improve the prognostic ozone in the troposphere, a simple relaxation-based solution may be an alternative. This could be achieved either by modifying the coefficients of the linear scheme, or by adding a separate relaxation to a (preferably latitude-longitude-height resolved) climatology of tropospheric ozone derived from the CAMS system. Although a monthly-mean climatology cannot account for the variability of tropospheric ozone, it might be adequate for use in the radiation scheme.

To address the shortcomings in the upper Stratosphere and Mesosphere, an alternative strategy could be considered. The assessments performed to date suggest that the mean state towards which the linear model relaxes is inconsistent with the observed mean state. This inconsistency could be resolved if coefficients were adjusted to ensure a better mean state than presently used. The linear timescale coefficients of either the Cariolle and BMS scheme could be used as the starting point of such an approach in the first instance.

The main advantage of this approach is that the scheme could regularly be updated to relax to the best available climatology, e.g. one derived from the CAMS reanalysis.

In contrast, the main limitation is that it requires a good observational climatology: while working well in regions where the CAMS reanalysis is driven by data, it might still be inadequate in the mesosphere where the ozone reanalyses are poorly constrained by observations. In this region, estimates from an external source (such as MIPAS; López-Puertas et al, 2018) may be required.

Any of these adaptations of the linear scheme will require careful testing in data assimilation mode with special attention to changes to the climatological and ensemble-based background error statistics. Recent experience with ERA5 showed issues in the ensemble runs because the ensemble-based calculation of background errors for O_3 can be related to specific characteristics of the ozone chemistry parametrisations.

Appendix C: AC schemes available in the IFS

Several schemes for AC have been implemented as part of the GEMS, MACC and CAMS developments and available in the IFS under dedicated switches (table 1). These schemes represent an asset for the IFS as they can facilitate testing the impact on meteorological fields of representing AC with different level of complexity. In this respect, the vertical resolution increase from 60 to 137 levels of the operational CAMS suite from Cy46r1 will be beneficial as consistent with the HRES one. An update from a T_L511 grid to a $T_{Co}511$ one, thus using a lower-resolution version of the HRES horizontal grid, could also be an advantage.

Table 1: Available AC schemes in the IFS and their use in CAMS and NWP operations (O) and research (R) mode.

Variables	Scheme (developed by)	Reference	CAMS	NWP
Ozone	Cariolle linearised ozone scheme, (Météo-France and ECMWF)	Cariolle and Teyssèdre, 2007	O, R	O, R
Ozone	BMS linearised ozone scheme, (Uni of Leeds and ECMWF)	Monge-Sanz et al, 2011		R
Tropospheric chemistry	CB05 (KNMI and ECMWF)	Huijnen et al., 2010; Flemming et al, 2015	O, R	
Aerosol bin scheme (DD, SS, OM, BC, SO_4)	LOA/LMDZ scheme (CNRS and ECMWF)	Morcrette et al, 2009	O, R	R
Aerosol modal scheme	GLOMAP (Uni of Leeds)	Mann et al, 2010	R	
CH_4/CO_2	GHG scheme (ECMWF)	Agustí-Panareda et al, 2014, 2016, 2017	O, R	
Stratospheric Chemistry	BASCOE (BIRA-IASB)	Errera et al., 2008	R	
Stratospheric and tropospheric chemistry	MOCAGE (Météo-France)	Peuch et al., 1999	R	
Stratospheric and tropospheric chemistry	MOZART (NCAR-UCAR) – not in IFS default cycle	Emmons et al. 2010	R	
Volcanic SO_2/SO_4 and test tracer	Volcanic point source (ECMWF)	Flemming and Inness (2013)	R	
Volcanic ash and SO_4	Volcanic point source (ECMWF)	Benedetti et al, 2011	R	
N_2O chemistry	Extracted from BASCOE (ECMWF)		R	
Radon	Extracted from CB05 (ECMWF)		R	
Flexible tracers	Fixed predefined lifetime and emissions (ECMWF)		R	

Appendix D: List of acronyms

Table 2: List of acronyms not defined in the text.

Acronym	Description
(3) 4D-Var	(Three)Four-dimensional variational data assimilation
C3S	Copernicus Climate Change Service
CALIOP	Cloud-Aerosol Lidar with Orthogonal Polarization
CALIPSO	Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation
CAMS	Copernicus Atmosphere Monitoring Service
CHE	CO2 Human Emission project
CHTESSEL	Carbon – Hydrology TESSEL
CMIP	Climate Modelling Inter-comparison Project
CH_4	Methane
CO	Carbon monoxide
CO_2	Carbon dioxide
COPE	Continuous Observation Processing Environment
CTESSEL	Carbon TESSEL
CTM	Chemistry Transport Model
EDA	Ensemble of Data Assimilation
EDGAR	Emission Database for Global Atmospheric Research
EUMETSAT	European Organization for the Exploitation of Meteorological Satellites
GCM	General Circulation Model
GEMS	Global and regional Earth-system Monitoring using Satellite and in-situ data
GEMS	Geostationary Environment Monitoring Spectrometer
GLOMAP	Global Model of Aerosol Processes
HCHO	Formaldehyde
IR	Infrared

MACC	Monitoring Atmospheric Composition and Climate
MetOp	Meteorological Operational satellite programme
MLS	Microwave Limb Sounder
MODIS	Moderate Resolution Imaging Spectroradiometer
NCEP	National Center for Enviromental Prediction
NRT	Near-real time
NWP	Numerical Weather Prediction
QBO	Quasi-Biennial Oscillation
RTTOV	Radiative Transfer for TOVS
SO_2	Sulphur dioxide
SODA	Satellite Ozone Data Assimilation
T_{Co}	Triangular-cubic-octahedral
TEMPO	Tropospheric Emissions: Monitoring of POllution
TESSEL	Tiled ECMWF Scheme for Surface Exchanges over Land
T_L	Triangular-linear
WMO	World Meteorological Office

References

Agustí-Panareda, A. and Co-authors, 2014: Forecasting global atmospheric CO_2, *Atmos. Chem. Phys.*, 14, 11959-11983, doi:10.5194/acp-14-11959-2014.

Agustí-Panareda, A., S. Massart, F. Chevallier, G. Balsamo, S. Boussetta, E. Dutra and A. Beljaars, 2016: A biogenic CO_2 flux adjustment scheme for the mitigation of large-scale biases in global atmospheric CO_2 analyses and forecasts. *Atmos. Chem. Phys.*, 16, 10399-10418, doi:10.5194/acp-16-10399-2016.

Agusti-Panareda, A., M. Diamantakis, V. Bayona, F. Klappenbach and A. Butz, 2017: Improving the inter-hemispheric gradient of total column atmospheric CO_2 and CH_4 in simulations with the ECMWF semi-Lagrangian atmospheric global model, *Geosci. Model Dev.*, 10, 1-18, doi: 10.5194/gmd-10-1-2017.

Aulignè, T, A.P. McNally and D.P. Dee, 2007: Adaptive bias correction for satellite data in a numerical weather prediction system. *Q.J.R. Meteorol. Soc*., 133: 631-642. doi:10.1002/qj.56.

Baklanov, A. and Co-authors, 2017: Key Issues for Seamless Integrated Chemistry–Meteorology Modeling. *Bull. Amer. Meteor. Soc*., 98, 2285–2292, https://doi.org/10.1175/BAMS-D-15-00166.1.

Benedetti, A. and M. Fisher, 2007: Background error statistics for aerosols, *Q.J R. Meteorol. Soc*., 133, 391–405.

Benedetti, A., and Co-authors, 2009: Aerosol analysis and forecast in the European Centre for Medium-Range Weather Forecasts Integrated Forecast System: 2. Data assimilation, *J. Geophys. Res. Atmos*., 114, D13205.

Benedetti, A., and Co-Authors, 2011: Simulations of volcanic plumes with the ECMWF/MACC aerosol system, *ECMWF Technical Memorandum* 653.

Benedetti, A. and F. Vitart, 2018: Can the Direct Effect of Aerosols Improve Subseasonal Predictability? Mon. Wea. Rev., 146, 3481–3498, https://doi.org/10.1175/MWR-D-17-0282.1.

Boussetta, S. and Co-authors, 2013: Natural land carbon dioxide exchanges in the ECMWF Integrated Forecasting System: Implementation and offline validation, *J. Geophys. Res*., DOI: 10.1002/jgrd.50488.

Bozzo, A., S. Remy, A. Benedetti, J. Flemming, P. Bechtold, M. Rodwell and J.-J. Morcrette, 2017: Implementation of a CAMS-based aerosol climatology in the IFS, *ECMWF Technical Memorandum* n. 801, ECMWF, Shinfield Park, Reading RG2-9AX, UK.

Buizza, R., E. Andersson, R. Forbes and M. Sleigh, 2017: The ECMWF Research to Operations (R2O) process. *ECMWF Technical Memorandum* n. 806 ECMWF, Shinfield Park, Reading RG2-9AX, UK, pp. 16.

Buizza, R. and Co-authors, 2018: The Development and Evaluation process followed at ECMWF to upgrade the Integrated Forecasting System (IFS), ECMWF/SAC/47(18)7 (item 6).

Cariolle, D. and H. Teyssèdre, 2007: A revised linear ozone photochemistry parameterization for use in transport and general circulation models: multi-annual simulations, *Atmos. Chem. Phys*., 7, 2183-2196.

Charlton, A.J., A. O'Neill, W.A. Lahoz and A.C. Massacand, 2004: Sensitivity of tropospheric forecasts to stratospheric initial conditions. *Q.J.R. Meteor. Soc.*, 130, 1771–1792.

Deconinck, W. and Co-authors, 2017: Atlas: A library for numerical weather prediction and climate modelling, *Computer Physics Communications*, 220, 188-204.

Dee, D.P., 2005: Bias and data assimilation. *Q.J.R. Meteorol. Soc*., 131: 3323-3343. doi:10.1256/qj.05.137.

Dethof, A. and Hólm, E. 2004: Ozone assimilation in the ERA-40 reanalysis project, *Q. J. Roy. Meteor. Soc*., 130, 2851–2872.

Dhomse and Co-authors, 2014: Aerosol microphysics simulations of the Mt. Pinatubo eruption with the UM-UKCA composition-climate model, *Atmos. Chem. Phys*., 14, 11221-11246, doi:10.5194/acp-14-11221-2014.

Diamantakis and Agusti-Panareda, 2017: A positive definite tracer mass fixer for high resolution weather and atmospheric composition forecasts, *ECMWF Technical Memorandum* n. 819, https://www.ecmwf.int/en/elibrary/17914-positive-definite-tracer-mass-fixer-high-resolution-weather-and-atmospheric.

Dragani, R., 2009: Variational bias correction of satellite ozone data. *Technical report* R43.8/RD/0934, ECMWF. Available from R. Dragani (rossana.dragani@ecmwf.int).

Dragani, R. and A.P. McNally, 2013: Operational assimilation of ozone-sensitive infrared radiances at ECMWF. *Q.J.R. Meteorol. Soc.*, 139: 2068-2080. doi:10.1002/qj.2106.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S., 2010: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geosci. Model Dev.*, 3, 43-67, https://doi.org/10.5194/gmd-3-43-2010.

Engelen, R.J., S. Serrar and F. Chevallier, 2009: Four-dimensional data assimilation of atmospheric CO2 using AIRS observations, *J. Geophys. Res.*, 114, D03303, doi:10.1029/2008JD010739.

Engelen, R.J., E. Andersson, F. Chevallier, A. Hollingsworth, M. Matricardi, A.P. McNally, J.-N. Thépaut and P.D. Watts, 2004: Estimating atmospheric CO_2 from advanced infrared satellite radiances within an operational 4D-Var data assimilation system: Methodology and first results, *J. Geophys. Res.*, 109, D19309, doi:10.1029/2004JD004777.

Engelen, R. J. and Bauer, P. 2011: The use of variable CO_2 in the data assimilation of AIRS and IASI radiances. *Q.J.R. Meteorol. Soc.*, 140: 958-965. doi:10.1002/qj.919.

Eresmaa, E., J. Letertre-Danczak, C. Lupu, N. Bormann and A.P. McNally, 2017: The assimilation of Cross-track Infrared Sounder radiances at ECMWF, *Q. J. R. Meteorol. Soc.*, 143, 3177-3188, doi:10.1002/qj.3171.

Errera, Q., and Co-Authors, 2008. 4D-Var assimilation of MIPAS chemical observations: ozone and nitrogen dioxide analyses, *Atmos. Chem. Phys.*, 8, 6169–6187, doi:10.5194/acp-8-6169-2008.

Eskes, H., 2003: Stratospheric ozone: satellite observations, data assimilation and forecasts. *Proceedings* of the 2003 ECMWF Annual seminar on Recent developments in data assimilation for atmosphere and ocean, available on-line at www.ecmwf.int/sites/default/files/elibrary/2003/9312-stratospheric-ozone-satellite-observations-data-assimilation-and-forecasts.pdf (last access on 8 June 2018).

Flemming, J., and A. Inness (2013), Volcanic sulfur dioxide plume forecasts based on UV satellite retrievals for the 2011 Grímsvötn and the 2010 Eyjafjallajökull eruption, *J. Geophys. Res. Atmos.*, 118, 10,172–10,189, doi: 10.1002/jgrd.50753.

Flemming, J., A. Inness, H. Flentje, V. Huijnen, P. Moinat, M.G. Schultz and Stein, O., 2009: Coupling global chemistry transport models to ECMWF's integrated forecast system, *Geosci. Model Dev.*, 2, 253-265, doi:10.5194/gmd-2-253-2009.

Flemming, J. and Co-authors, 2015: Tropospheric chemistry in the Integrated Forecasting System of ECMWF, *Geosci. Model Dev.*, 8, 975–1003, doi:10.5194/gmd-8-975-2015.

Flemming, J. and Co-authors, 2017: The CAMS interim Reanalysis of Carbon Monoxide, Ozone and Aerosol for 2003–2015, *Atmos. Chem. Phys*., 17, 1945-1983, doi:10.5194/acp-17-1945-2017.

Freitas and Co-authors, 2015: Evaluating aerosols impacts on numerical weather prediction: a WGNE exercise. In *Proceedings* of the Symposium on Coupled Chemistry-Meteorology/Climate Modelling, 23-25 February 2015.

Haywood, J., 2009: The strategy for aerosols and dust in climate, weather and air quality forecasting. *Met Office MOSAC-14*, Paper No. 14.11, available online at http://research.metoffice.gov.uk/research/nwp/publications/mosac/doc-2009-11.pdf, (last access 18 July 2018).

Hogan, R and Co-authors, 2017: Radiation in numerical weather prediction, *ECMWF Technical Memorandum* n. 816.

Hollingsworth, A. and Co-authors, 2008: Toward a Monitoring and Forecasting System For Atmospheric Composition: The GEMS Project. *Bull. Amer. Meteor. Soc*., 89, 1147–1164, doi:10.1175/2008BAMS2355.1.

Holben, B.N.,and Co-authors, 2001: An emerging ground-based aerosol climatology: Aerosol Optical Depth from AERONET, *J. Geophys. Res*., 106, 12 067-12 097.

Hólm, E.V., A. Untch, A. Simmons, R. Saunders, F. Bouttier and E. Andersson, 1999: Multivariate ozone assimilation in four-dimensional data assimilation. In *Proceedings* of SODA workshop on Chemical Data Assimilation, Publication 188, KNMI, De Bilt, the Netherlands.

Huijnen, V., and Co-Authors (2010). The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0, *Geosci. Model Dev*., 3, 445-473, doi:10.5194/gmd-3-445-2010.

Inness, A. and Co-authors.: 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.

IPCC (Intergovernmental Panel on Climate Change), Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp, doi:10.1017/CBO9781107415324, 2013.

Ivy, D.J., S. Solomon, D. Kinnison, M.J. Mills, A. Schmidt and R.R. Neely III, 2017: The influence of the Calbuco eruption on the 2015 Antarctic ozone hole in a fully coupled chemistry-climate model, *Geophys. Res. Lett*., 44, 2556–2561, doi: 10.1002/2016GL071925.

Janssens-Maenhout, G. and Coauthors, 2017: EDGAR v4.3.2 Global Atlas of the three major Greenhouse Gas Emissions for the period 1970–2012, *Earth Syst. Sci. Data Discuss.*, doi: 10.5194/essd-2017-79.

Kühnlein, C. and P.K. Smolarkiewicz, 2017: An unstructured-mesh finite-volume MPDATA for compressible atmospheric dynamics, *Journal of Computational Physics*, 334, 16-30, doi: 10.1016/j.jcp.2016.12.054.

Leblanc, T. and I. S. McDermid, 2001: Quasi-biennial oscillation signatures in ozone and temperature observed by lidar at Mauna Loa, Hawaii (19.5°N, 155.6°W), *J. Geophys. Res.*, 106(D14), 14869–14874, doi: 10.1029/2001JD900162.

López-Puertas, M. and Co-authors, 2018: MIPAS observations of ozone in the middle atmosphere, *Atmos. Meas. Tech.*, 11(4), 2187–2212.

Lupu, C. and A.P. McNally, 2013: Wind tracing with ozone-sensitive radiances from SEVIRI, EUMETSAT/ECMWF Fellowship Programme, Research Report No 31.

Mann, G.W. and Co-authors, 2010: Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the UKCA composition-climate model, *Geosci. Model Dev.*, 3, 519-551, https://doi.org/10.5194/gmd-3-519-2010.

Massart, S. and Co-authors, 2016: Ability of the 4-D-Var analysis of the GOSAT BESD XCO2 retrievals to characterize atmospheric CO2 at large and synoptic scales, *Atmos. Chem. Phys.*, 16, 1653-1671, doi:10.5194/acp-16-1653-2016.

Matricardi, M. and A.P. McNally, 2014: The direct assimilation of principal components of IASI spectra in the ECMWF 4D-Var. *Q.J.R. Meteorol. Soc.*, 140: 573-582. doi:10.1002/qj.2156.

McNally, A.P and M. Vesperini, 1996: Variational analysis of humidity information from TOVS radiances, *Q.J.R. Meteorol. Soc.*, 122, 1521–1544.

Monge-Sanz, B.M., M.P. Chipperfield, D. Cariolle and W. Feng, 2011: Results from a new linear O3 scheme with embedded heterogeneous chemistry compared with the parent full-chemistry 3-D CTM, *Atmos. Chem. Phys.*, 11, 1227-1242.

Morcrette, J.-J., and Co-authors (2009). Aerosol analysis and forecast in the ECMWF Integrated Forecast System. Forward modelling, *J. Geophys. Res.*, 114, D06206, doi:10.1029/2008JD011235.

Mulcahy, J. P., D. N. Walters, N. Bellouin, and S. F. Milton (2014). Impacts of increasing the aerosol complexity in the Met Office global numerical weather prediction model, *Atmos. Chem. Phys.*, 14, 4749–4778, 2014, doi:10.5194/acp-14-4749-2014

Parrish, D.F. and J.C. Derber (1992), The National Meteorological Center's spectral statistical-interpolation analysis system, *Mon. Weather Rev.*, 120, 1747–1763.

Peubey, C. and A.P. McNally, 2009: Characterization of the impact of geostationary clear-sky radiances on wind analyses in a 4D-Var context. *Q.J.R. Meteorol. Soc.*, 135, 1865–1876, doi: 10.1002/qj.500.

Peuch, V.-H. and Co-Authors, 1999. MOCAGE: Modèle de Chimie-Transport à Grande Echelle, Acte de l'Atelier de Modélisation de l'Atmosphère, 1999, 33-36.

Reale O., K.M. Lau and A. Da Silva, 2011: Impact of an Interactive Aerosol on the African Easterly Jet in the NASA GEOS-5 Global Forecasting System. *Wea. Forecasting*, 26, 504-519.

Rodwell, M.J. and T. Jung, 2008: Understanding the local and global impacts of model physics changes: an aerosol example. *Q.J.R. Meteorol. Soc*., 134: 1479-1497. doi:10.1002/qj.298.

Schroedter-Homscheidt, M., A. Oumbe, A. Benedetti and J.-J. Morcrette, 2013: Aerosols for concentrating solar electricity production forecasts: Requirement quantification and ECMWF/MACC aerosol forecast assessment. *Bull. Amer. Meteor. Soc*., 94, 903–914.

Semane, N. and Co-authors, 2009: On the extraction of wind information from the assimilation of ozone profiles in Météo-France 4-D-Var operational NWP suite. *Atmos. Chem. Phys*., 9, 4855–4867.

Smolarkiewicz, P.K., W. Deconinck, M. Hamrud, C. Kühnlein, G. Mozdzynski, J. Szmelter and N.P. Wedi, 2016: A finite-volume module for simulating global all-scale atmospheric flows, *Journal of Computational Physics*, 314, 287-304, doi:10.1016/j.jcp.2016.03.015.

Spracklen, DV; Pringle, KJ; Carslaw, KS; Chipperfield, MP; Mann, GW, 2005. A global off-line model of size-resolved aerosol microphysics: I. Model development and prediction of aerosol properties, *Atmos. Chem. Phys.*, 5, pp2227-2252.

Stockdale and Co-authors, 2018: SEAS5 and the future evolution of long range forecast systems, ECMWF/SAC/47(18)12 (item 10.2).

Struthers, H., R. Brugge, W.A. Lahoz, A. O'Neill and R. Swinbank, 2002: Assimilation of Ozone Profiles and Total Column Measurements into a Global General Circulation Model, *J. Geophys. Res*., 107, doi:10.1029/2001JD000957.

Tavolato, C., and L. Isaksen, 2014: On the use of a Huber norm for observation quality control in the ECMWF 4D-Var, *ECMWF Technical Memorandum* n. 744.

Tegen, I., P. Hollrig, M. Chin, I. Fung, D. Jacob and J. Penner, 1997: Contribution of different aerosol species to the global aerosol extinction optical thickness: Estimates from model results, *J. Geophys. Res.*, 102(D20), 23895–23915, doi: 10.1029/97JD01864.

Váňa, F., P. Düben, S. Lang, T. Palmer, M. Leutbecher, D. Salmond, and G. Carver, 2017: Single Precision in Weather Forecasting Models: An Evaluation with the IFS. *Mon. Wea. Rev*., 145, 495–502, https://doi.org/10.1175/MWR-D-16-0228.1.

Ziemke, J.R., S. Chandra, G.J. Labow, P.K. Bhartia, L. Froidevaux and J.C. Witte.: A global climatology of tropospheric and stratospheric ozone derived from Aura OMI and MLS measurements, *Atmos. Chem. Phys*., 11, 9237-9251, https://doi.org/10.5194/acp-11-9237-2011, 2011.