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Aerosol optical depth calculations using the Bulgarian Chemical Weather Forecast System

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Abstract: Five different methods are tested for estimating the aerosol optical depth using results from the Bulgarian Chemical Weather Forecast System. Four of the methods are embedded in the chemical transport model of the system; the fifth one (FlexAOD) is adapted from a post-processing tool, developed for global chemistry models. The results of the five approaches are discussed qualitatively, showing maps for AOD spatial distribution over Europe for a selected day. The performance of the code FlexAOD with results from BgCWFS is discussed for a period of four days in March 2018, characterized with Saharan Dust outbreak. The preliminary evaluation with AOD from the Copernicus based forecast system CAMS-ECMWF and with data from AERONET stations shows that BgCWFS underestimates AOD and suggest further developments of the system with assimilation of satellite derived data.

Keywords: AOD, Bulgarian Chemical Weather Forecast System,

1. INTRODUCTION

Aerosols play an important role in the radiative budget of the atmosphere – they have direct effects through scattering and absorption of radiation, as well as indirect effects through mechanisms influencing the cloud formation (IPCC, 2013). For their key role in the global climate, numerous studies have been undertaken in the past decades in order to improve their representation in the global climate models (e.g. Mallet et al., 2017). Aerosol particles in the atmosphere are also linked to long range atmospheric transport

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processes (sand dust storms, wild fires, volcanic ash), and to regional anthropogenic pollution, and thus have significant impact on air quality and human health.

Aerosol optical depth (AOD) is a measure of the column-integrated extinction of radiation, it is approximately proportional to the aerosol mass concentration and is one of the primary climate change indicators (Sullivan et al, 2017). The aerosol distribution is highly inhomogeneous in space and time, as it is linked to different natural and anthropogenic sources, complex microphysical processes and transformations the particles undergo during the atmospheric transport. This leads to high uncertainty in the estimation of the aerosol's effects on the Earth's atmosphere (Boucher et al., 2013) and triggers numerous investigations both at global and regional scales using different methods – measurements, modelling, or combination of both.

AOD is routinely obtained by the global surface based network AERONET (AERosol RObotic NETwork, (https://aeronet.gsfc.nasa.gov), as well as by spaceborn remote sensing instruments, e.g. MODIS- Moderate Resolution Imaging Spectroradiometer aboard the NASA (National Aeronautics and Space Administration) Terra and Aqua platforms (Hyer et al., 2011).

Global and regional chemical transport models (CTM) are powerful tools to estimate aerosol optical properties with high temporal and spatial resolution, however they exhibit uncertainties related to, among others, model's parameterizations and emissions input, (Park et al, 2011). To improve the accuracy of modeled aerosols, data assimilation of satellite retrieved data has been exploited in the last years, both for forecasting purposes (Benedetti et al., 2009) and for analyzing aerosol processes in different geographical areas (e.g. Colarco et al., 2010; Wang et al., 2017).

The Bulgarian Chemical Weather Forecast System (BgCWFS) is running operationally at the National Institute of Meteorology and Hydrology (NIMH) since 2012. The system provides 72h forecast for key pollutants – ozone, nitrogen dioxide and particulate matter over five different geographical areas – from European scale- down to the city scale of Sofia with results presented on the web (http://info.meteo.bg/cw2.1/ and http://info.meteo.bg/cw2.2/). As part of continuous improvement efforts, and in the framework of a recent ESA supported project SIDUAQ (http://space.bas.bg/SIDUAQ/), BgCWFS is tested for assimilation of satellite retrieved AOD data for the five domains. This requires extension of the current version of the system with modules for AOD calculations.

Here, we present and discuss some preliminary results for different AOD calculation methods for use in BgCWFS. Section 2 outlines the main modules of the system and the methodology of 5 different AOD algorithms. Section 3 presents results for selected days of March 2018 characterized by Saharan Dust outbreaks towards Bulgaria. The AOD spatial distribution simulated by BgCWFS is compared to modelling results from the Copernicus Atmosphere Monitoring Service (CAMS-ECMWF) forecast (https://apps.ecmwf.int/datasets/data/cams-nrealtime/levtype=sfc/).

2. MODEL DESCRIPTION AND METHODS

2.1. Overview of the modeling system

The Bulgarian Chemical Weather Forecasting System (BgCWFS), designed on the base of US EPA Models-3 air quality modelling system, has the following computational modules:

- WRF v.3.6.1 - Weather Research and Forecasting Model as meteorological preprocessor, (Skamarock and Klemp, 2008);
- CMAQ v.4.6 - Community Multi-scale Air Quality model, an Eulerian Chemical Transport Model, (Byun and Schere, 2006);
- MCIP v.3.6 - Meteorology-Chemistry Interface Processor;
- SMOKE v.2.4 - Sparse Matrix Operator Kernel Emissions Modelling System - the emission pre-processor to CMAQ (used partly- for calculating biogenic emissions and for merging emission files for area sources (AS), large point sources (LPS), and biogenic sources).

BgCWFS is set up for simulations in 5 nested domains - Europe (81 km resolution), Balkan Peninsula (27 km), Bulgaria (9 km), Sofia-district (3 km) and Sofia-city (1 km).

WRF is driven by data from the National Centers for Environmental Prediction Global Forecast System (NCEP GFS) with space resolution of 1°× 1°, and temporal resolution of 6-h. Initial conditions for CMAQ are part of previous day calculations. Boundary conditions for CMAQ at the European domain are from predefined in the model vertical concentration profiles, all other domains receive their boundary conditions from the previous one in the hierarchy. The emissions are based on TNO inventory for 2009 (Kuenen et al. 2014), for Bulgaria national emission inventories for 2010 are used. The chemical mechanism is "cb4_ae4_aq", the output from the chemical modelling produces hourly 3D files containing concentrations, depositions and visibility parameters for 78 pollutants: 52 gaseous and 26 aerosol species divided in 3 modes – Aitken mode ($d < 1$ μm), Accumulation mode ($1\ \mu m < d < 2.5\ \mu m$) and Coarse mode ($2.5\ \mu m < d < 10\ \mu m$).

More details on BgCWFS can be found in Syrakov et al., 2012, 2013a,b, 2014, 2016 and Gadzhev et al., 2012.

2.2. AOD calculations

CMAQ does not calculate directly AOD, but uses calculated profiles of the aerosol species for its estimation. The vertical structure of CMAQ consists of fourteen σ-levels with varying thickness, the Planetary Boundary Layer (PBL) is presented by the lowest 8 levels, the top of the model is at about 20 km a.g.l.

Here, we used 5 different methods to calculate AOD at wavelength 550 nm and the respective AOD are denoted hereafter as *AOD_Mie, AOD_Rec, AOD_Imp, AOD_Rev, and AOT_C.*

2.2.1. CMAQ AERO module: AOD_Mie and AOD_Rec

CMAQ is able to determine the reduction in the visibility caused by the presence of particulate matter (PM), perceived as haze. The module AERO calculates four visibility indices – aerosol optical extinction coefficients (EXT) and a parameter defined by the extinction coefficient, called *deciview.*

The AERO module includes two methods for EXT calculation: the Mie method and the reconstruction method. The Mie method is based on Mie scattering (a generalized particulate light-scattering mechanism that follows from the laws of electromagnetism applied to particulate matter) over the entire range of particle sizes to obtain visibility parameters for each model grid cell at each time step. This method does not require aerosol chemical composition information. Because routine measurements of aerosol species mass concentrations are often available, but particle size distribution information is not, an additional method of calculating extinction has also been included in AERO module. This is an empirical approach known as reconstructed extinction. The method, explained in Malm et al. (1994), uses the mass concentrations of aerosol chemical compositions to calculate the total extinction coefficient for the wavelength of 550 nm with a lookup table. For this method also data for the relative humidity are needed. More detailed descriptions of the PM calculation techniques used in CMAQ can be found in Binkowski and Shankar (1995), Binkowski and Roselle (2003), and Byun and Schere (2006).

Tang et al. (2017) point out that the reconstruction method, due to its simplicity and convenience is widely used not only for CMAQ aerosols module, but also for converting observed aerosol mass concentrations to extinction coefficient. The Mie method, based on aerosol physical characteristics, is relatively hard to use in data assimilations as the data assimilations target the mass concentrations of aerosol compositions, not the aerosol physical characteristics directly.

Originally CMAQ calculates two extinction coefficients (*EXT_Mie and EXT_Rec)* only at surface layer. As far as AOD can be determined as vertical integral of EXT, modifications in the CMAQ source code were made in order to calculate and output the visibility parameters at each model layer i with thickness DZ_i, and thus to estimate AOD using:

$$AOD = \sum_{i=1}^{Nz} EXT_i \times DZ_i \,, N_z - number\ of\ model\ layers \tag{1}$$

Thus, from the CMAQ AERO module two different values of AOD were simulated – *AOD_Mie* and *AOD_Rec.*

2.2.2. IMPROVE methodology: AOD_Imp, AOD_Rev

The Interagency Monitoring of Protected Visual Environments (IMPROVE), is a particle monitoring network established in the USA in 1985 (http://vista.cira.colostate.

edu/Improve/improve-program/). It consists of approximately 160 sites at which fine particulate matter (PM2.5) mass and major species concentrations and coarse particulate matter (PM10) mass concentrations are determined. On the base of serious amount of measurements of visibility parameters and aerosol concentrations, a simple algorithm (called further *original*) was elaborated to estimate light extinction from the measured species concentrations in 1999. Later, a *revised* algorithm was developed that is more consistent with the recent atmospheric aerosol literature and reduces bias for high and low light extinction extremes. The *revised* algorithm differs from the *original* algorithm mainly by a term for estimating sea salt light scattering from Cl^- ion data, by using site-specific Rayleigh scattering, and by employing a split component extinction efficiency associated with large and small size mode sulfate, nitrate and organic mass species (Pitchford et al, 2007).

For the calculation of AOD by this methodology, specific scripts and Fortran programs were elaborated in order to extract 8 groups of aerosols required by the IMPROVE algorithms from the CMAQ's concentration output. The groups of aerosols are ORG (Organic mass), FIN (Fine soil), CRS (Coarse mass), LAC (Light absorbing carbon), SLF (Sulphates, $SO4^-$), NTR (Nitrates, $NO3^-$), AMO (Amonium, $NH4^+$) and SAL (Sea Salt). They are extracted as linear combinations between the original CMAQ's aerosol species as follows:

ORG = AORGAJ+ AORGAI+ 1.167 *(AORGPAJ+AORGPAI)+AORGBJ+AORGBI
FIN = A25J
CRS = ACORS+ASOIL
LAC = AECJ+AECI
SLF = ASO4J+ASO4I
NTR = ANO3J+ANO3I
SAL = ANAJ+ACLJ+ANAK+ACLK+ASO4K,

where the indices "I" ,"J" and "K" denote Aitken, Accumulation and Coarse mode aerosol. AORGA denotes anthropogenic secondary organic mass, AORGPA – the primary organic mass, AORGB – the secondary biogenic organic mass, and A25 – the unspecified anthropogenic mass.

For both of the CMAQ embedded approaches for extinction coefficients, the relative humidity (RH) is needed at all levels and grid points. RH is not a direct output by MCIP, thus, a program was elaborated to calculate RH based on the other 3D meteorological variables. RH is calculated from the vapor pressure and saturated vapor pressure using:

$$RH = 100.\frac{e}{e_s}$$

where e is estimated using air density, ambient temperature and water vapor mixing ratio, and e_s is estimated using the Magnus formula.

The values for AOD calculated with this methodology are noted as *AOD_Imp* (*original*) and *AOD_Rev (revised)*.

2.2.3. The FlexAOD tool

The Flexible Aerosol Optical Depth (FlexAOD) post-processing tool was originally developed to calculate aerosol optical properties for GEOS-Chem global modelling system (http://acmg.seas.harvard.edu/geos/index.html) and is described in the CEOS-Chem wiki-site (http://wiki.seas.harvard.edu/geos-chem/index.php/FlexAOD). Further on it was modified by Gabriele Curci (University of L'Aquila, IT, http://pumpkin.aquila.infn.it/flexaod/) to calculate off-line hourly AOD from the CMAQ model archived hourly three-dimensional, speciated aerosols (i.e., sulfate, nitrate, ammonium, black carbon (BC), Organic carbon (OC), sea salt, soil dust) and meteorological fields (RH), Curci et al. (2014), Curci et al. (2019), Jin et al. (2019).

Under the assumption of spherical particles, aerosol optical properties are calculated based on Mie theory. Given the size distributions for each aerosol species, the aerosol light extinction (EXT_k) at a given model layer k is calculated as follows, Curci, (2012):

$$EXT_k = \sum_{i=1}^{N_a} (3/4)\, Q_{e,dry,i} \times f\left(RH_{k,i}\right) \times M_{i,k} / (r_{e,dry,i}\ \rho_i) \qquad k = 1, N_z \qquad (2)$$

where i refers to the species, Na is the number of aerosol species (Na =5: sulfate–nitrate–ammonium (SNA), OC, BC, dust, sea salt), $Q_{e,dry,i}$ is the Mie extinction efficiency of species i averaged over the dry size distribution, $f\,(RH_{k,i})$ is the hygroscopic growth factor of species i at given RH_k, ρ_i is the aerosol density of species i, $M_{i,k}$ is the aerosol mass of species i at layer k, and $r_{e,dry,i}$ is the dry effective radius. AOD is then calculated as the vertical integral of EXT_k across all model layers after (1).

In its present version, FlexAOD allows the user to:

- calculate AOD at several wavelengths (550 nm used in our case) without repeating the chemical simulation;
- calculate additional aerosol optical properties (e.g. single scattering albedo, backscattering coefficient, asymmetry factor, etc.);
- extract aerosol concentrations and optical profiles at selected locations and times.

FlexAOD produces two kinds of outputs – in *grads* and *netcdf* -formats. Here, the *netcdf* output is exploited. As far as it contains several variables, only this one interesting for us is extracted. It is called *AOT_C* (Atmospheric Optical Thickness, Columnar).

2.3. Case selection

The demonstration of AOD simulated by BgSWFS can be performed for any day of the year but we decided to choice a period, that will be tested afterwards also for assimilation of satellite retrieved AOD, i.e. a period with good density of satellite measurements over the territory of the Balkan Peninsula and particularly Bulgaria. The case period is 20-27 March 2018, when a Saharan Dust Storm was approaching from south-west towards Bulgaria. The satellite derived aerosol absorbing index (AAI) from GOME 2 instrument clearly shows the Saharan Dust outbreak in the Eastern Mediterranean,

Figure 1. The model CMAQ possess a built-in module for dust emissions calculations, however in our model set-up it is useless for such events because Sahara is out of the BgCWFS biggest domain (Europe). In cases when Saharan dust is transported towards Bulgaria, the assimilation of AOD from satellites would cause an increase of the aerosol concentrations and would improve modelled aerosols. The overpass hour of the *MetOp* satellite over Bulgaria is between 08 and 09 UTC each day, so we decided to assign the data to a common passing hour of 09:00 UTC.

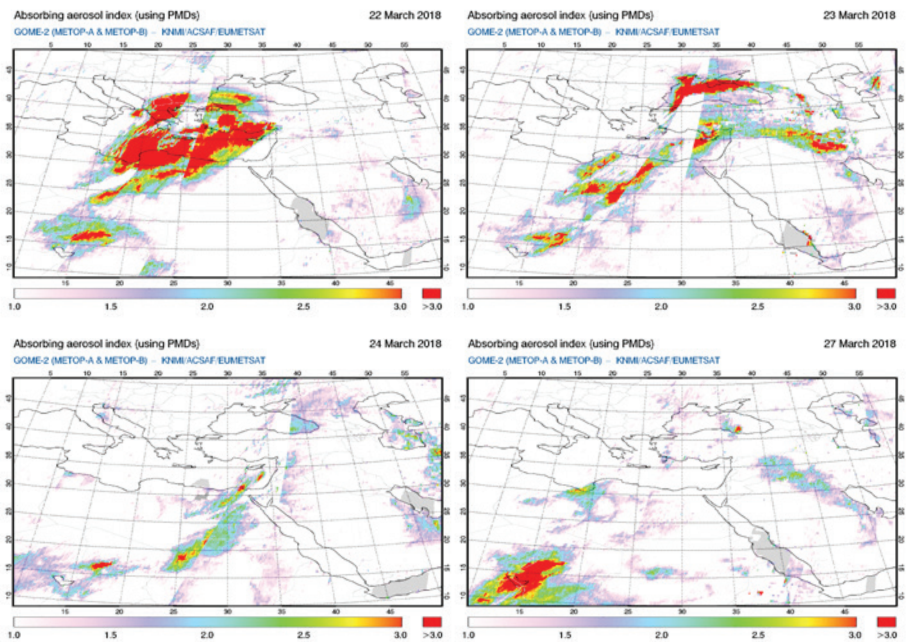


Fig. 1. AAI in the Eastern Mediterranean from GOME 2 for selected days of the chosen period 20-27.03.2018 (source: http://sacs.aeronomie.be/nrt/index.php)

2.4. Evaluation methods

The evaluation of model estimated AOD is performed usually based on comparison to observational data (satellite retrieved, or from the ground based aerosol robotic network AERONET (Holben et al., 1998)) considering long-time periods – at least monthly values, but more often on seasonal or yearly basis. Another possibility is comparison to estimates by other models. For example the model inter-comparison initiative AQMEII-3 (Palacios-Peña et al., 2019) focussed on AOD over Europe in 2010.

There are numerous models focussing particularly on the prediction of mineral dust events and simulating the dust AOD. These activities are led by the World Meteorological Organization (WMO) with the implementation of the Sand and Dust Storm Warning Advisory and Assessment System (SDS-WAS) (http://www.wmo.int/sdswas). At the

Regional Node for Northern Africa, Middle East and Europe (NAMEE), hosted by the State Meteorological Agency of Spain (AEMET) and the Barcelona Supercomputing Center (BSC), more than 10 different models provide operational dust forecasts for NAMEE (https://sds-was.aemet.es/). Some of these models are built on dust emissions from arid and semi-arid areas and on dust cycle schemes only, and do not consider anthropogenic emissions. For the purposes in this study we have selected one of the modelling systems (CAMS-ECMWF), which takes into account also anthropogenic emissions and simulates AOD due to various size of particles, in different bins of the fine and coarse fraction.

Hourly values for AOD are provided by the modelling system Copernicus Atmosphere Monitoring Service at the European Centre for Medium-Range Weather Forecasts (CAMS–ECMWF). This comprehensive system, running operationally at global level, assimilates AOD satellite data (Benedetti et al., 2009).

Table 1. AERONET stations used for the case study

station_name	Site_Latitude(Degrees)	Site_Longitude(Degrees)
Brno_airport (CZ)	49.1565	16.6833
HohenpeissenbergDWD (GE)	47.8019	11.0119
Iasi_LOASL (RO)	47.1931	27.5556
Ispra (IT)	45.8031	8.6267
Lampedusa (IT)	35.5167	12.6317
Strzyzow (PL)	49.8786	21.8613
Venise (IT)	45.3139	12.5083
Vienna_BOKU (AT)	48.2379	16.3316

Here we use CAMS-ECMWF forecasted hourly values for AOD at 550 nm, downloading the dataset over Europe with grid resolution (0.125° × 0.125°) and for the time step at 09:00 UTC from https://apps.ecmwf.int/datasets/data/cams-nrealtime/levtype=sfc/. These data are used for qualitative comparison of AOD spatial distribution over Europe (maps).

Some quantitative evaluation is provided for the AERONET stations (Table 1), which have data in the case study period. The time window used for retrieving AOD at a single station is from 8:30 UTC to 09:30 UTC, and the mean value in this interval is further used for comparisons.

The evaluation of AOD calculated by BgCWFS is not an objective of this study, as it requires additional simulations and is planned as separate activity in the frame of the SIDUAQ project. Thus, the comparisons provided here are very limited aiming just to demonstrate availability of AOD estimates from different sources in the case study period.

3. RESULTS AND DISCUSSION

The AOD spatial distribution by the different methods in BgCWFS is shown in Figures 2, 3, and 4 for 22.03.2018 09:00 UTC.

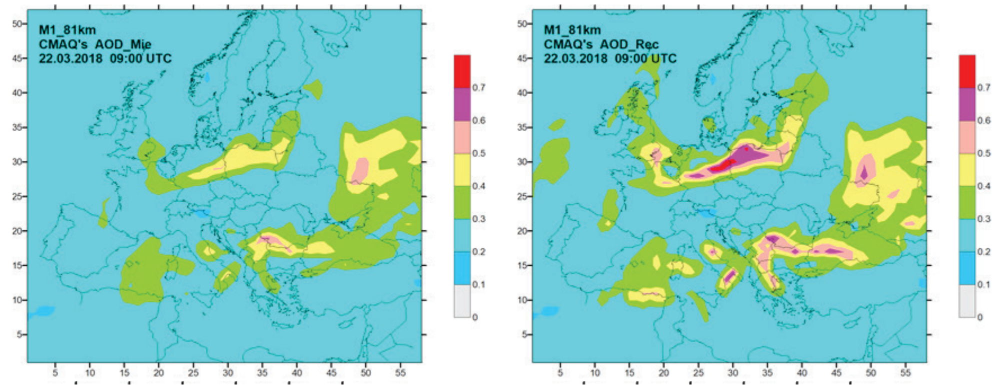


Fig. 2. CMAQ's estimates for AOD for the EU domain of BgCWFS: AOD_Mie (left) and AOD_Rec (right)

One can notice that the spatial distribution for both embedded methods in CMAQ has similar configurations, with AOD by Mie method producing higher values than the IMPROVE method, and with *AOD_Rec* showing higher maximal values.

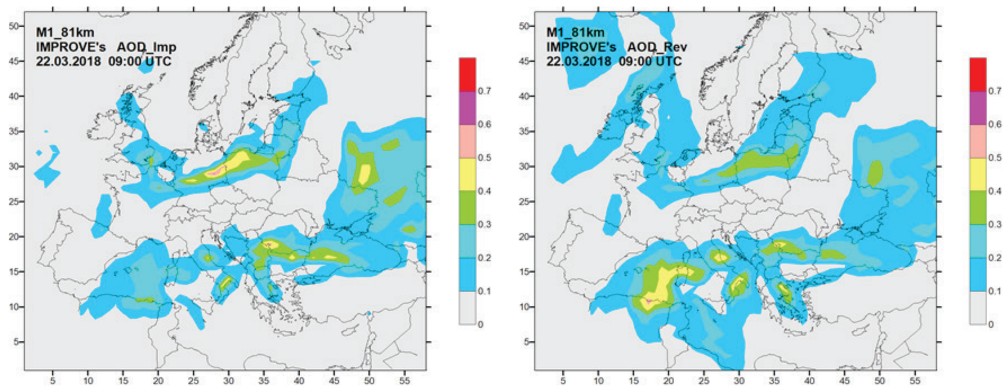


Fig. 3. IMPROVE based AOD for the EU domain of BgCWFS, AOD_Imp (left) and AOD_Rev (right)

The AOD spatial distribution by the FlexAOD method (Figure 4) has rather different pattern, although the regions with higher AOD roughly match the regions simulated by the other methods (southern Italy, Northern Africa). At the same time there are notable differences in both the spatial extension and the AOD magnitude (e.g. northwest of Bulgaria and in the eastern part of the domain).

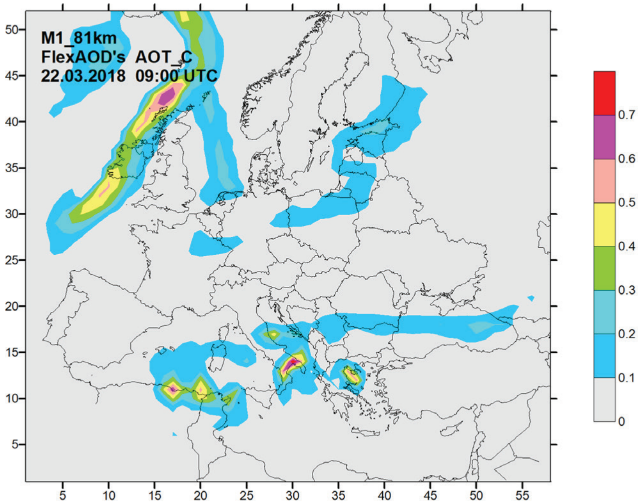


Fig. 4. FlexAOD produced AOT_C for the EU domain of BgCWFS.

It is not a trivial task to estimate which of the above AOD methods performs best. In the literature one can find preferences of different methods by different authors. Curci et al. (2014) apply both FlexAOD and CMAQ's AOD_Mie methods. Lee et al. (2011) use AOD_Rec for assimilating satellite measurements, Park et al. (2011) and Tang et al. (2017) use the same method with small modifications. Roy et al. (2007) use the *original* IMPROVE algorithm for calculating AOD, while Pour-Biazar et al. (2011) use the *revised* IMPROVE methodology. Wang et al. (2017) use both IMPROVE algorithms. FlexAOD is used by Jin et al. (2017) and Curci et al. (2019). Thus, each one of these five methods is applied by the modelling community and has its value.

At this preliminary stage, we will focus more on FlexAOD as it is used in the frame of the global atmospheric chemistry model GEOS_Chem for assimilation of satellite data and incorporates the scientific achievements in the field of optics of atmospheric aerosols. To note that it allows to calculate AOD at various wavelengths, and thus gives more possibilities for characterization of different aerosol types.

The AOD spatial distribution in Europe from BgCWFS with FlexAOD is compared to AOD estimated by CAMS forecast for 22, 23, 24 and 27 March 2018 at 09:00 UTC (Figure 5). As the CAMS-ECMWF system assimilates satellite AOD, it is able to capture the Saharan Dust outbreak, on the contrary the BgCWFS does not take Saharan dust emissions into account, so the main differences are south of Bulgaria. Common for both models in the spatial distribution are regions with elevated aerosols, not necessary related to the Saharan outbreak: on 22.03.18 - the higher levels over the north-western part of UK and Northern Africa, on 23.03 – elevated values in the central Mediterranean and north of France, on 24.03 – higher values north of Spain.

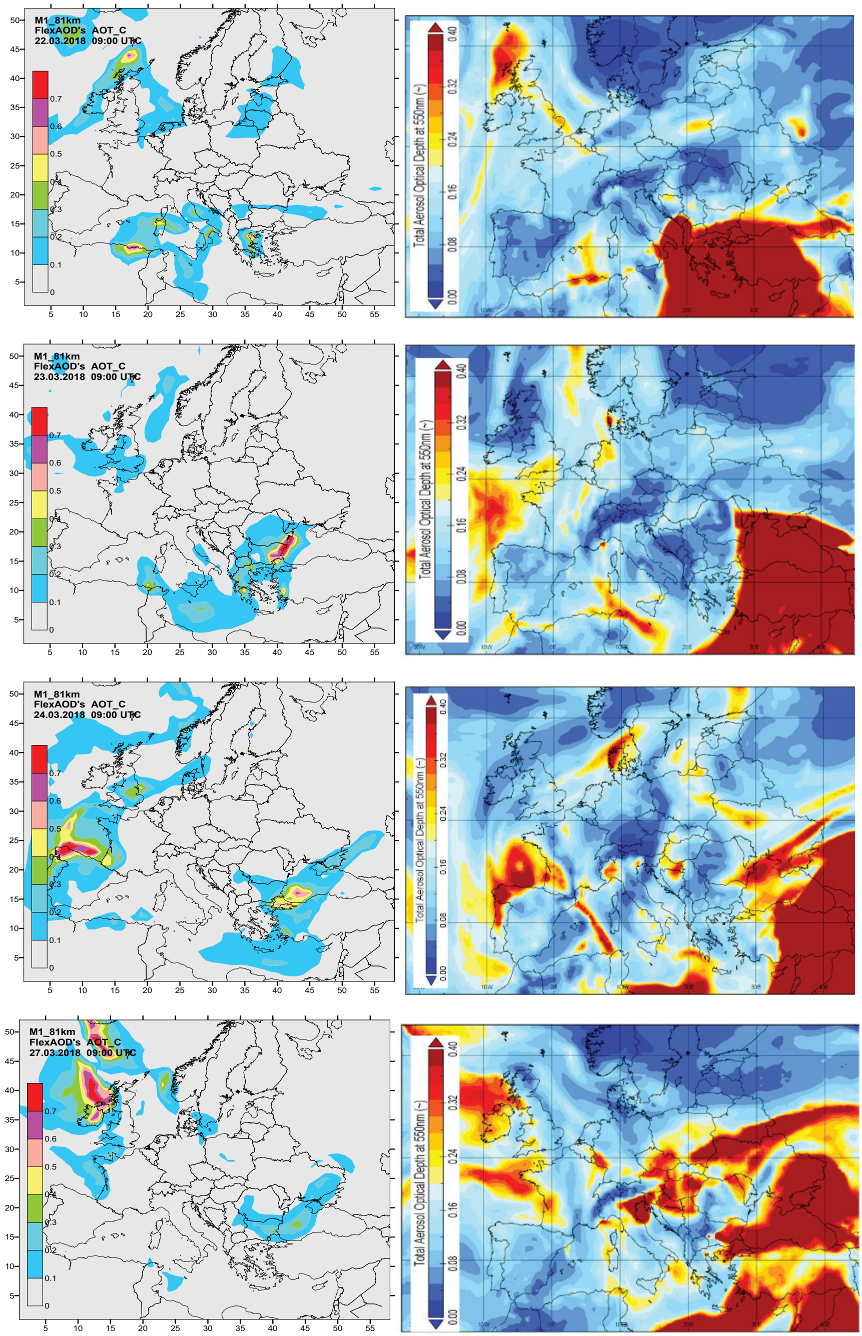
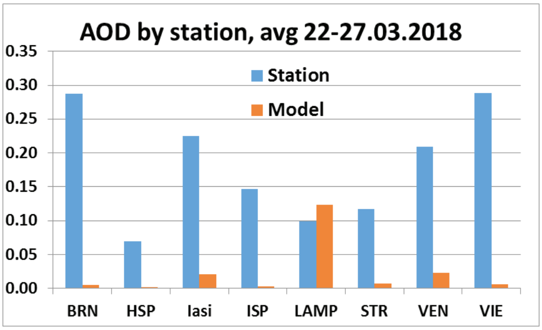


Fig. 5. AOD calculated over Europe for 22., 23., 24. and 27.03.2018 at 09:00 UTC by BgCFWS with FlexAOD (left) and by CAMS-ECMWFS (right), Generated using Copernicus Atmosphere Monitoring Service Information [2019].

The comparison to AOD from the AERONET stations is based on rather limited number of observations for the case period in this study. The number of paired (model and station) is 20, moreover, the comparison should be treated with caution as the model grid resolution is rather rough (81 km). The AOD from the stations is at 500 nm.

Figure 6 shows the comparison of calculated AOD to stations' AOD. For each day of the case period the model underestimates AOD by a factor of 5 (on average). The underestimation is significant at the selected stations, with only one exception – for the site Lampedusa (IT). The discrepancies between model and observations is strongly influenced by stations 'representation errors – both on the temporal and spatial scale, and for some sites it can reach 100% (Schutgens, 2019)



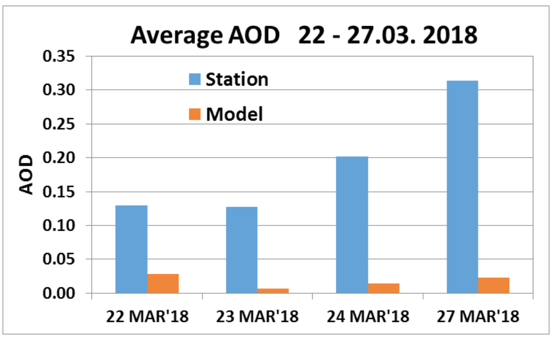


Fig. 6. Comparison of model AOD to stations's AOD: AOD averaged for the selected period on single station (top); average for all stations on single day (botom)

Different factors might be responsible for the AOD underestimation by BgCWFS – emissions, model set-up, aerosol's parameterizations etc. To recall that the emissions used in the system are for 2009, while the measurements represent actual conditions. Thus, part of the underestimation is probably due to outdated emissions data in BgCWFS. However, looking at the AOD spatial distribution in Figure 5, it seems that the most important factor for the underestimation in the southern part of the domain is the lack of dust emissions from the region of Sahara. The period was characterized by a Saharan Dust outbreak towards Bulgaria, but the biggest domain in BgCWFS does not include the Saharan area, and thus no emissions of mineral dusts are included. The CAMS-

ECMWFS model very clearly indicates a zone with higher AOD over the south-eastern part of the domain. The AAI satellite pictures in Figure 1 also show the advancement of Saharan Dust Storm in the period of this case study.

This underestimation suggests that assimilation of AOD from satellites may improve the performance of BgCWFS for such cases. The investigation on this will be object of another study.

4. SUMMARY

Five different methods and estimates for AOD in the Bulgarian Weather Forecast System are studied, coded and tested for demonstration purposes. Based on the limited data for their comparison and on the experience by different authors from the literature, it is impossible to firmly state which one preforms better. A method based on the FlexAOD code is selected for further analysis in BgCWFS, as the code offers more flexibility options in estimating the AOD. Preliminary evaluation with AOD from the forecast system CMAS-ECMWF system and from AERONET stations show that BgCWFS underestimates AOD for the studied period, characterized with Saharan Dust outbreak. The assimilation of satellite retrieved AOD could be beneficial for model results in such cases, but additional simulations and analysis are necessary. Further steps in this direction are related to model simulations for at least one-month period, and to evaluation of model performance (without – and with satellite data assimilation) looking also at particulate matter concentrations at ground level.

5. ACKNOWLEDGMENTS

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