

Response to the Referees

“A complex aerosol transport event over Europe during the 2017 Storm Ophelia in CAMS forecast systems: analysis and evaluation”

by Dimitris Akritidis et al.

Dear Dr. Kazadzis

We would like to thank the three Referees for their constructive comments and their time devoted on reviewing our manuscript, contributing to its improvement.

On the following pages, we present our point by point response (blue colour) to the comments raised by the Referees (black colour), along with the corresponding changes made in the manuscript. A version of the revised manuscript with our changes highlighted (blue and red) is also included.

Sincerely,

Dimitris Akritidis (on behalf of all the co-authors)

Anonymous Referee #1

We would like to thank Reviewer #1 for her/his time devoted and the constructive and helpful comments to which we will respond point by point.

This is a very well, and concisely written evaluation of the ability of the CAMS global and regional systems for forecasting a rather particular aerosol event, being a combination of high loadings of dust, biomass burning and sea salt particles. I find interesting to learn that the IFS forecasts excluding data-assimilation already show similar performance as the configuration including data-assimilation. Does this either point at a very good forecast model strength, or rather at deficiencies in the data assimilation setup?

We thank the Reviewer for the general comment. In fact, the use of data assimilation improves the performance of CAMS system in AOD forecast, yet it seems to leave relatively unaffected the PM₁₀ and PM_{2.5} forecast near the surface. Both speculations seem valid. On the one hand, IFS is a state-of-the-art model that apparently reproduces the aerosol transport and the induced increase in particulate matter (PM) surface concentrations, without the use of data assimilation. On the other hand, it seems that the data assimilation of a columnar aerosol product (AOD) has no substantial effect on surface PM air quality forecast, for the examined event.

I find it only unfortunate that authors only assess the first-day forecasts. This only a limited view of the forecast capability of the system is presented. It would have been very interesting if the authors would have shown the (likely decay in) forecast capability for the second and third day forecasts, as this would give a better handle for 'citizens and policy-makers' on the reliability of forecasts on a time scale where they are able to take action, which is one of the key objectives of this study. Is it possible for authors to make statements on this?

Apart from this, I only have some small comments.

We agree with the suggestion raised by the Reviewer and thus in the Revised Manuscript (RM) we have included IFS and RegEns day-2 and day-3 forecasts of PM₁₀ and PM_{2.5} in Figures 9, 10, S2 and S3. Additionally, we have extended Figure 11 including the results from day-2 and day-3 forecasts. Accordingly, we have modified/added several parts in the RM.

The following paragraph is included in the RM at P11, L21-29:

“The capability of IFS and RegEns systems to forecast the observed PM₁₀ and PM_{2.5} surface concentrations two and three days in advance, is finally discussed. As depicted in Figure 9, IFS day-2 and day-3 forecasts reproduce the distinct increases in observed PM₁₀ surface concentrations exhibiting similar FGE values but lower correlation scores (in most of the stations) compared to day-1 forecast (Fig. 11c and d). The same applies in the case of PM_{2.5} (Fig. 10), except that the correlation scores for IFS day-2 and day-3 forecasts are not systematically lower than that of day-1 forecast (Fig. 11c and d). As regards the RegEns, although it fairly predicts the observed peaks in PM₁₀ and PM_{2.5} up to three days in advance (Fig. 9 and 10), there is a systematic deterioration of its performance in terms of temporal variability over forecast

time. More specifically, the correlation coefficient decreases from day-1 to day-2 forecast and from day-2 to day-3 forecast for almost all examined stations (Fig. 11e)."

The following sentence is included in the Conclusions of the RM at P12, L25-27:

"A deterioration of the RegEns forecast performance is found over forecast time for both PM₁₀ and PM_{2.5}, characterized by a decrease of the correlation coefficient for the vast majority of the examined stations, which is partially seen in IFS for the case of PM₁₀."

We have modified a part of the Abstract, see P1, L15-17 at the RM:

"Regarding the footprint on air quality, both CAMS global and regional forecast systems are generally able to reproduce the observed signal of increase in surface particulate matter concentrations. The regional component performs better in terms of bias and temporal variability, with the correlation deteriorating over forecast time."

Page 4, l 10L 'day-1 forecasts'. Are these the forecasts initiated at 0h00? Just because the CAMS operational system provides currently two forecasts per day.

Yes. Both CAMS global and regional forecasts used in the paper are initiated at 00:00 UTC. In the RM, we have included the respective information at P4, L8 and P5, L12 as follows: **"(initiated at 00:00Z)"**.

Page 4. The authors provide empirical formulae to compute the PM₁₀ and PM_{2.5} concentrations from the global system, which are crucial for the definition of the contribution of aerosol to the air quality statistics. These equations appear rather empirical, and also different depending on the model version. Can the authors give some more background information as to how these metrics are designed? Also, to what extent does the definition of this metric contribute to differences seen in Figure 8?

These are indeed empirical formulae which are updated when necessary with IFS cycle updates. Regarding the PM₁₀ and PM_{2.5} formulae, the factors applied in sea salt bins are used to transform sea salt from 80% relative humidity ambient conditions to dry, while the rest correspond to the fraction of each aerosol type included in PM₁₀ and PM_{2.5}. More information on the description and evaluation of the aerosol scheme used in IFS is provided by Remy et al. (2019) which we have included in the RM at P4, L18-19 as follows: **"A detailed description and evaluation of the aerosol scheme used in IFS can be found in Remy et al. (2019)."** The differences seen in Figure 8 are likely also due to the definition of PM₁₀ and PM_{2.5} in IFS and CAMS regional models, which is already stated in the manuscript as **"The aforementioned inconsistencies are likely due to the different definition of PM₁₀ and PM_{2.5} in IFS and each CAMS regional air quality model,.."** (see P10, L12-13 of the RM). A quantitative estimation of this is beyond the scope of this paper, yet that would be an interesting task for a future study.

Page 5, l3: "CAMS regional models assimilate PM₁₀ and PM_{2.5}" : does this hold for all CAMS regional models, or only for some? Also, are PM₁₀ and PM_{2.5} the modeled tracer fields in the regional models, or are they computed from underlying aerosol composition fields, as is the case

for the global model? In summary, could the authors elaborate a little more on how PM is modeled in the regional system?

Indeed, not all CAMS regional models were assimilating PM₁₀ and PM_{2.5} at the time of the event. More specifically, CHIMERE and EURAD were assimilating both PM₁₀ and PM_{2.5}, MOCAGE only PM₁₀, while SILAM and MATCH were assimilating only PM_{2.5}. PM₁₀ and PM_{2.5} in the regional models are calculated using underlying aerosol species, as it is the case with the CAMS global model. However, each model is free to choose how exactly this is done, as the aerosol schemes and thus the aerosol species of each model differ. These details have been added in the RM at P5, L3-8: **“Several CAMS regional models assimilate PM₁₀ and PM_{2.5} surface observations from various stations of the EEA’s (European Environment Agency) Air Quality e-reporting database, but not satellite aerosol products. More specifically, during the period of interest (October 2017), CHIMERE and EURAD were assimilating both PM₁₀ and PM_{2.5}, MOCAGE only PM₁₀ and, finally, SILAM and MATCH were assimilating only PM_{2.5}. PM₁₀ and PM_{2.5} concentrations in the regional models are calculated using simulated aerosol fields specific to each regional model.”**

page 7, line 26: “reducing the bias”-> reducing the error

Done.

page 8, line 1: “the percentage” -> the modeled percentage

Done.

page 10, line 30: “implementation”: I would rather write “application”.

Done.

page 11, line 9: “The CAMS regional system seems to better predict the : :.” : why not write something like: “for this event the CAMS regional system shows better ..”

We have included the phrase **“For the examined event,”** in the beginning of the respective sentence (P12, L20).

References

Rémy, S., Kipling, Z., Flemming, J., Boucher, O., Nabat, P., Michou, M., Bozzo, A., Ades, M., Huijnen, V., Benedetti, A., Engelen, R., Peuch, V.-H., and Morcrette, J.-J.: Description and evaluation of the tropospheric aerosol scheme in the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecasting System (IFS-AER, cycle 45R1), *Geoscientific Model Development*, 12, 4627–4659, <https://doi.org/10.5194/gmd-12-4627-2019>, 2019

Anonymous Referee #2

We would like to thank Reviewer #2 for her/his time devoted and the constructive and helpful comments to which we will respond point by point.

The authors analyse an interesting meteorological/aerosol event named Ophelia, that took place in mid-October 2017 over western Europe. They use CAMS global day-1 forecast data to assess the aerosol spatiotemporal distribution and determine the source and type of aerosol species. Validating the performance of CAMS in these kind of events is quite crucial to understand the current limitations in CAMS aerosol forecast system and the possible upgrades that need to be planned (e.g. assimilating more and different kinds of aerosol observations). The manuscript is well written providing all the necessary information to introduce the event to the reader. I would recommend for publication in ACP after addressing some minor comments below:

We thank the Reviewer for the general comment.

CAMS AOD is evaluated using dependent observations (as mentioned at P7, L10-11) from MODIS Dark Target and Deep Blue algorithms showing reasonable agreement with the observations and improvement in both correlation and bias in comparison to the no assimilation experiment. The paper does not provide a comparison with independent observations to prove that data assimilation improves AOD. I would suggest to mention again in conclusions that this is a dependent evaluation (P10, L28-31) or perform a small analysis using independent observations (e.g. AERONET AOD) to prove that CAMS AOD really improves, although the latter option may be out of the scope of this paper.

We agree with the comment raised from the Reviewer and thus in the Conclusions of the RM (P12, L8-9) we explicitly state that findings are drawn from a depended evaluation: **“The dependent evaluation against MODIS satellite observations reveals a satisfactory agreement with CAMS global AOD₅₅₀ (R=0.77 and FGE=0.4), while the comparison..”**. As assumed by the Reviewer, a comparison with AERONET AOD was not a primary scope of the present paper. Moreover, there is a limited availability of AERONET data during the passage of Storm Ophelia (due to cloud presence), with a few stations exhibiting fragments of observations (see also Figure 9 in the study of Osborne et al. (2019)), which in any case are not sufficient to conduct a comprehensive independent evaluation of CAMS global AOD product taking also into account its temporal resolution (3-hour).

Figure 6 and discussion starting at P8, L1: A very interesting analysis highlighting the contribution of each species to AOD. Nevertheless the contribution of each species to AOD may differ depending on the aerosol-species optical properties. Black carbon AOD is low (and Absorption AOD high) in comparison to the other species. A 15% to 25% contribution of black carbon AOD to the total AOD might seem mediocre, but climatologically is only observed during fire season in the Tropical band. Maybe the authors would like to comment on that.

We agree with the comment and therefore we include the following sentence in the RM (P9, L3-5): **“Such black carbon contributions are considered high being similar to climatological contributions over global fire hot spots (e.g. summertime central southern Africa; Penning de Vries et al. (2015)).”**

Figure 7 and discussion starting at P8, L17: Although this is more of a qualitative comparison to discuss the vertical distribution of aerosols per species, the depiction of CALIPSO and IFS are very different above 4km in both tracks. Is it possible to conclude something about the observations or the model? For example is CALIPSO unable to retrieve trustworthy measurements during the pass of Ophelia above 4km or is IFS overestimating aerosol mixing ratio above a certain height? The respective Figure has been revised correcting a minor bug in our code and converting IFS sea salt from 80% RH ambient conditions to dry, as suggested by ECMWF, which was inadvertently omitted. However, the Figure and the respective discussion have not changed substantially. Regarding the inconsistencies above 4km, this is a rather complex and still uncertain issue. As discussed comprehensively in Georgoulas et al. (2018) the non-zero aerosol concentrations appearing systematically at heights above 5 km in model simulations compared to remote sensing observations (also see Mona et al., 2014; Biniotoglou et al., 2015; Cuevas et al., 2015 and Ansmann et al., 2017) could be due to:

- The way the model deals with the aerosol distribution in different size bins and aerosol deposition, vertical transport and mixing.
- The IFS assimilation process and the fact that nitrate aerosols are missing in the present IFS aerosol version, which might lead to the appearance of small aerosol concentrations further high.
- The limitation of CALIPSO in detecting aerosol layers with signals lower than the satellite's signal-to-noise ratio.

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Anonymous Referee #3

We would like to thank Reviewer #3 for her/his time devoted and the constructive and helpful comments to which we will respond point by point.

The authors analyse a remarkable event of combined desert dust and biomass burning aerosol transport over Europe using observations and model results, and in the process evaluate some aspects of the CAMS air quality forecast system. Studying such events is important because of their quite drastic impact on the European air quality, potential implications on clouds and weather, and impairment of solar energy yield. Their distinct air quality signatures are good benchmarks for forecast systems. The subject is relevant and within the scope of ACP (there is some overlap with the GMD scope regarding the model evaluation aspect), the article is well organised and written so that I recommend publication after addressing the following comments:

We thank the Reviewer for the general comment.

Page 1, line 11: Throughout the manuscript you use the correlation (coefficient) r , here you present the shared variance (and I assume you are referring to r^2). Because the term is less common or even used with different meaning, and to be consistent with the main text, I suggest to stick to r .

We agree with the comment and thus we have replaced “shared variance of 60%” with “correlation coefficient of 0.77” in the Revised Manuscript (RM) P1, L11.

Page 1, line 13: Please expand IFS here.

Done.

Section 2.1: Some more information about the aerosol representation in the model would be helpful: does the model assume the different aerosol types to be externally mixed, both regarding the aerosol optical properties and regarding any chemical or physical interactions between different components? This would be interesting to know especially since the event under consideration involves dust, biomass burning aerosol and sea salt.

We agree with the comment raised by the Reviewer. Indeed, the IFS aerosol types are treated as externally mixed (separate particles). The following has been included in the RM P4, L7: “The different IFS aerosol types are treated as externally mixed (Inness et al., 2019a).”

Page 4, line 8 and 9: The units should be micro metres not metres and it has to be mentioned that the numbers represent radii not diameters. What bins are used for the other aerosol types, e.g., what is BC1 vs. BC2?

We thank the Reviewer for the comment, to provide a more explanatory description. We have replaced “size bins” with “radius size bins” (m were already replaced with μm in the discussion version of the manuscript). The two bins used for organic matter and black carbon aerosols stand for hydrophobic and hydrophilic. We have modified the respective discussion in the RM P4, L15-18 as follows: “where ρ the air density, SS1 the sea salt radius size bin 1 (0.03-0.5 μm), SS2 the

sea salt radius size bin 2 (0.5-5 μm), DD1 the desert dust radius size bin 1 (0.03–0.55 μm), DD2 the desert dust radius size bin 2 (0.55–0.9 μm), DD3 the desert dust radius size bin 3 (0.9–20 μm), OM1 the hydrophobic organic matter, OM2 the hydrophilic organic matter, BC1 the hydrophobic black carbon, BC2 the hydrophilic black carbon and SU1 the aerosol sulfate (ECMWF, 2020).

Page 4, Eqs. (1) and (2): I understand these equations follow recommendations elsewhere, but could you indicate in the text where the factors come from? Also, SS3 is not considered for PM_{2.5} (and PM₁₀), because the radius is > 5 μm , but according to the intervals provided above, not considering DD3 for PM_{2.5} seems to ignore dust particles between 0.9 and 1.25 μm radius.

The factors applied in sea salt bins are used to transform sea salt from 80% relative humidity ambient conditions to dry, while the rest correspond to the fraction of each aerosol type included in PM₁₀ and PM_{2.5}. More information on the description and evaluation of the aerosol scheme used in IFS is provided by Remy et al. (2019) which we have included in the RM P4, L18-19 as follows: **“A detailed description and evaluation of the aerosol scheme used in IFS can be found in Remy et al. (2019).”** As for the disregarded DD3 radius size range 0.9-1.25 μm in PM_{2.5}, this is a limitation of the definition.

Page 5, line 10: Please expand LT once
Done. See P5, L16 of the RM.

Page 5, lines 28ff: Which selection criteria for the stations were used exactly?

Since here we examine the aerosol transport during Ophelia and the associated impacts on air quality in CAMS forecast systems, we only consider rural EMEP stations that fulfill the following criteria:

1. They are located over western Europe and away from the aerosol sources (i.e. Northern Africa and Iberian Peninsula).
2. They lie across the plumes of high AOD loadings during the examined event.
3. They exhibit remarkable increases in PM₁₀ and PM_{2.5} surface concentrations (visual inspection).

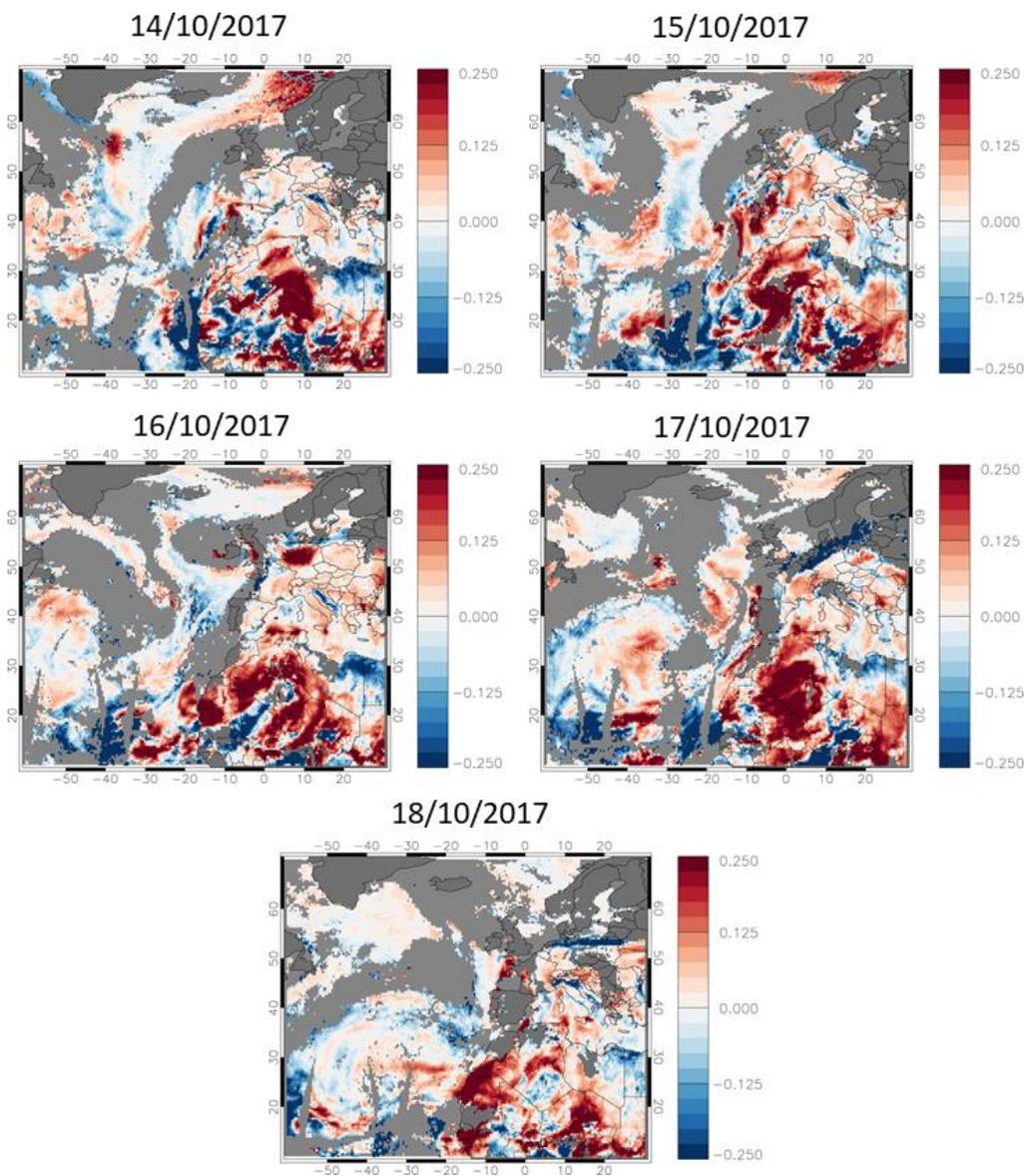
Accordingly, we have replaced the sentence **“The stations are located over the broader western European areas where dust and biomass burning transport occurred”** with the following in the RM P6, L2-4: **“The stations are located over western Europe and away from the dust and biomass burning sources, lie across the plumes of high AOD loadings, exhibiting significant increases in PM₁₀ and PM_{2.5} surface concentrations during the examined event.”**

Page 7, line 26: The FGE deserves to be introduced by an equation, moreover uppercase is more common.

We have added a new subsection (2.4 Statistical metrics) in the RM, where we present basic information and formulae for the Pearson correlation coefficient and the fractional gross error. We have also replaced all instances of fge with FGE following the suggestion in both text and figures of the RM.

Page 7, lines 30 to 34: How can this conclusion be aligned with Fig. 4, where in many regions with low AOD the AOD is enhance by data assimilation?

What we mean is that generally over areas with low “observed” AOD the IFS forecast tends to overestimate and vice versa. This agrees with the comment raised by the Reviewer, as over several areas with low observed AOD the IFS (with DA) forecast mostly overestimates. We have replaced “**low AOD values**” with “**low observed AOD values**” in the RM (P8, L26) for clarity. In support of the above we present here the differences between IFS AOD₅₅₀ and MODIS/Terra and Aqua AOD₅₅₀, which along with the left column of Figure 4 (MODIS/Terra and Aqua AOD₅₅₀ fields) confirms our conclusion.



Figs. 9 and 10: I find the similarity of the PM10 and PM2.5 composition surprising, normally I would expect a higher dust and sea salt fraction in PM10 than in PM2.5. Does this indicate some

limitation of the model? After all, from Eqs. (1) and (2) it is clear that the contributions in Fig. 9 and 10 cannot be that different, and the largest particles (SS3 and DD3) are not relevant at all, but is that realistic? Do the stations provide the PM_{2.5} and/or PM₁₀ composition for comparison, or is there any other suitable data source to validate this aspect?

Indeed, as depicted from the PM₁₀ and PM_{2.5} equations the contributions are similar. This was also the case in some sensitivity calculations we performed using random values for each aerosol type and bin. The equations themselves may play a role in that. The applied formulae are mostly empirical and thus we agree that may insert uncertainties (e.g. large particles are underrepresented). However, it should be also considered that in enhanced dust and sea salt conditions the total PM₁₀ and PM_{2.5} concentrations are also higher likely resulting in similar relative contributions in PM₁₀ and PM_{2.5}. Unfortunately, from the examined stations only GB0048R and GB1055R EMEP stations provide fragments of PM₁₀ and PM_{2.5} composition data for the examined period. These data are not sufficient for a comprehensive estimation of dust and sea salt contribution to PM₁₀ and PM_{2.5} due to missing data and lack of all PM₁₀ and PM_{2.5} aerosol components (available with gaps: NH₄⁺, Ca⁺⁺, Cl⁻, Mg⁺⁺, NO₃⁻, K⁺, Na⁺ and SO₄⁻). From data for the GB1055R station during the 16-17 October PM peak, it seems that although the individual dust and sea salt component concentrations are higher in PM₁₀ compared to PM_{2.5}, it is the total PM₁₀ concentrations that are also higher compared to total PM_{2.5} that probably result in similar contributions.

Generally, I believe links in footnotes and within the text are supposed to be moved to the References section to comply with the journal standards.

Done. In the RM we have replaced all footnotes and links with References according to the Journal standards.

References

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A complex aerosol transport event over Europe during the 2017 Storm Ophelia in CAMS forecast systems: analysis and evaluation

Dimitris Akritidis¹, Eleni Katragkou¹, Aristeidis K. Georgoulas¹, Prodromos Zanis¹, Stergios Kartsios¹, Johannes Flemming², Antje Inness², John Douros³, and Henk Eskes³

¹Department of Meteorology and Climatology, School of Geology, Aristotle University of Thessaloniki, Thessaloniki, Greece

²European Centre for Medium-Range Weather Forecasts (ECMWF), Reading, UK

³Royal Netherlands Meteorological Institute (KNMI), De Bilt, the Netherlands

Correspondence: D. Akritidis (dakritid@geo.auth.gr)

Abstract. In mid-October 2017 Storm Ophelia crossed over western coastal Europe, inducing the combined transport of Saharan dust and Iberian biomass burning aerosols over several European areas. In this study we assess the performance of the Copernicus Atmosphere Monitoring Service (CAMS) forecast systems during this complex aerosol transport event, and the potential benefits that data assimilation and regional models could bring. To this end, CAMS global and regional ~~day-1~~ forecast data are analyzed and compared against observations from passive (MODIS: Moderate resolution Imaging Spectroradiometer aboard Terra and Aqua) and active (CALIOP/CALIPSO: Cloud-Aerosol Lidar with Orthogonal Polarization aboard Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) ~~satellites~~ satellite sensors, and ground-based measurements (EMEP: European Monitoring and Evaluation Programme). The analysis of CAMS global forecast indicates that dust and smoke aerosols, discretely located on the warm and cold front of Ophelia, respectively, are affecting the aerosol atmospheric composition over Europe during the passage of the Storm. The observed MODIS Aerosol Optical Depth (AOD) values are satisfactorily reproduced by CAMS global forecast system, with a ~~shared variance of 60%~~ correlation coefficient of 0.77 and a fractional gross error (~~fge~~ FGE) of 0.4. The comparison with a CAMS global control simulation not including data assimilation, indicates a significant improvement in the bias due to data assimilation implementation, as the ~~fge~~ FGE decreases by 32%. The qualitative evaluation of the IFS (Integrated Forecast System) dominant aerosol type and location against the CALIPSO observations overall reveals a good agreement. Regarding the footprint on air quality, both CAMS global and regional forecast systems are generally able to reproduce the observed signal of increase in surface particulate matter concentrations, ~~with the regional component performing-~~ The regional component performs better in terms of bias and temporal variability, with the correlation deteriorating over forecast time. Yet, both products exhibit inconsistencies on the quantitative and temporal representation of the observed surface particulate matter enhancements, stressing the need for further development of the air quality forecast systems, for even more accurate and timely support of citizens and policy-makers.

1 Introduction

Atmospheric aerosols play a prominent role in atmospheric composition, climate and human health (Pöschl, 2005; IPCC, 2013). Given the broad variety of their natural and anthropogenic sources, their relatively short lifetime, and their different formation

mechanisms, aerosols exhibit highly variable spatiotemporal distribution around the globe (Putaud et al., 2010; Boucher, 2015). Over Europe, apart from local emissions, particulate matter quantities are also determined by atmospheric transport through mesoscale weather systems (Ansmann et al., 2003; Kallos et al., 2007; Pey et al., 2013), occasionally implying significant implications for air quality and public health (Stafoggia et al., 2016). Consequently, the operational forecast of atmospheric composition is essential in the direction of supporting social and health policy-makers.

Aerosols interact with solar radiation directly through scattering and absorption (Haywood and Boucher, 2000), and indirectly by modifying the micro- and macro-physical properties of clouds (Lohmann and Feichter, 2005), modulating the energy balance in the earth-atmosphere system, and by extension the climate. More specifically, they may act as cloud condensation nuclei (CCN) and ice nuclei (IN) increasing the cloud albedo under constant water paths (Twomey, 1974), as well as cloud lifetime and coverage suppressing the precipitation formation (Albrecht, 1989). In addition, the presence of absorbing aerosols alters the thermodynamic state of the atmosphere (semi-direct effect) affecting clouds and precipitation in various ways (Koch and Del Genio, 2010).

Exposure to particulate matter air pollution is well known to have significant implications to respiratory- and cardiovascular-based mortality (Russell and Brunekreef, 2009; Lelieveld et al., 2015; Burnett et al., 2018). Fine particulate matter with a diameter less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) has more profound effects on human health compared to coarse particulate matter with a diameter less than $10 \mu\text{m}$ (PM_{10}), as smaller particles can penetrate more efficiently into the lungs and the indoor environments, while they can also be transported over greater distances given their smaller mass and therefore their longer lifetime (Pope and Dockery, 2006). Recently, Lelieveld et al. (2019) based on new hazard ratio functions and ambient air pollution exposure data estimated that air pollution reduces the mean life expectancy by more than 2 years in Europe. Moreover, aerosols can pose significant hazards for aviation, occasionally resulting in flight delays and cancellations, as intense episodic dust and sea salt events can alter visibility (Gertisser, 2010; Tighe, 2015), while the presence of dust and volcanic particles may potentially cause aircraft engine damage (Gertisser, 2010). The impact of aerosols on photovoltaic generation is mainly due to reduction of surface solar radiation through scattering and absorption (Li et al., 2017), and deposition of dust on solar panels reducing their energy production potential (Beattie et al., 2012).

North African dust is the dominant source of mineral dust in the atmosphere (Ginoux et al., 2012), affecting the air quality of Europe from the Mediterranean (Querol et al., 2009; Georgoulias et al., 2016) up to Scandinavia (Ansmann et al., 2003). The main transport pathways of Saharan dust towards Europe are either passing through the Mediterranean by northward flow associated with the presence of cyclones/anticyclones westward/eastward of a Saharan dust source, or via the Atlantic route including a westward transport of Saharan dust by trade winds over the Atlantic followed by northward and then eastward flow towards Europe (Israelevich et al., 2012). Over Europe, biomass burning emissions from wildfires are a present air pollution risk, likely to increase in a changing climate (Knorr et al., 2017; Turco et al., 2017). The regional scale (Amiridis et al., 2009) and intercontinental (Markowicz et al., 2016) transport of smoke over Europe may affect the atmospheric composition and the local air quality (Sciare et al., 2008).

During the recent years various global and regional integrated forecast systems incorporating the online coupling between meteorology and atmospheric composition have been developed to support research, forecast and policy activities (Baklanov

et al., 2014). Copernicus¹ ([Copernicus, 2020](https://www.copernicus.eu)) is the European system for monitoring Earth coordinated by the European Union. The Copernicus Atmosphere Monitoring Service (CAMS)¹ ([CAMS, 2020a](https://www.cams2020a.eu)) is one of the six services that Copernicus provides, using a comprehensive global assimilation and forecasting system to assess the state and composition of the atmosphere on a daily basis. It incorporates information from models and observations, providing daily 5-day forecasts of atmospheric composition fields, such as chemically reactive gases and aerosols (Flemming et al., 2015; Inness et al., 2015). The CAMS global modeling system is also used to provide the boundary conditions for the CAMS ensemble of regional air quality models, which produce 4-day forecasts of European air quality at higher horizontal resolution. To increase confidence of operational use, unravel limitations and further improve the CAMS forecast systems, analysis and evaluation of its performance during complex and challenging situations is deemed necessary.

10 On the morning of October 16th 2017 a red sky phenomenon was reported on several sites of the United Kingdom (UK) (Telegraph, 2017), as a result of the high aerosol loadings in the overlying atmosphere (Harrison et al., 2018; Osborne et al., 2019), while similar reports on the morning of October 17th were given for the Netherlands as well (NLTIMES, 2017). These high aerosol loadings were the result of transport of desert dust from Northern Africa and smoke from Iberian wildfires. This study aims to analyze and evaluate the performance of CAMS global and regional forecast systems during this challenging combined dust and biomass burning transport event over western Europe induced from the passage of Storm Ophelia in mid-15 October 2017. To this end, CAMS global and regional day-1 forecast data are used, along with ground-based observations (EMEP: European Monitoring and Evaluation Programme), and observations from passive (MODIS: Moderate resolution Imaging Spectroradiometer aboard Terra and Aqua) and active (CALIOP/CALIPSO: Cloud-Aerosol Lidar with Orthogonal Polarization/Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite remote sensors. Furthermore, the impact of data assimilation on the CAMS global aerosol burden representation is also explored using a control simulation without the use of data assimilation. [Finally, to explore the forecast capability over time, CAMS global and regional day-2 and day-3 forecasts of PM₁₀ and PM_{2.5} concentrations are also analyzed.](#) This paper builds upon the work of Akritidis et al. (2018), assessing the performance of CAMS forecast products, the potential benefits of data assimilation in CAMS global system and the added value that CAMS regional models could bring, during a complex aerosol transport event. The structure of the paper is as follows. Section 2 presents the CAMS forecast systems and the observational data that are used to validate their performance. 25 Section 3 shows the analysis and evaluation results, and Section 4 summarises the key findings.

2 CAMS forecast systems~~and~~, validation data [and metrics](#)

2.1 CAMS global forecast system

One of the core CAMS products is the 5-day global chemical weather forecast, based on the European Centre for Medium-30 Range Weather Forecasts (ECMWF) Integrated Forecasting System (IFS). In October 2017, the forecast data were products of the IFS model cycle 43r3 having a horizontal resolution of about 40 km and 60 vertical levels reaching up to 0.1 hPa.

¹ <https://www.copernicus.eu>

The IFS modeling system uses an extended version of the Carbon Bond 2005 (CB05) chemical mechanism (Flemming et al., 2015) and the Morcrette et al. (2009) aerosol parameterization. Several chemical species, including ozone, nitrogen dioxide, carbon monoxide, sulfur dioxide are assimilated using products of several satellite missions (Inness et al., 2015, 2019b, and references therein). Moreover, the IFS assimilates total Aerosol Optical Depth (AOD) retrievals from the MODIS instrument on NASA's Aqua and Terra satellite, as well as PMAp (Polare Multi Sensor Aerosol product) aerosol retrievals over sea, a combined GOME-2/AVHRR product produced by Eumetsat (EUMETSAT, 2020), and subsequently updates the individual aerosol components based on their fractional contribution to the total aerosol mass (Benedetti et al., 2009; Georgoulias et al., 2018). Five types of aerosols are included, namely sea salt, dust, hydrophilic and hydrophobic organic matter and black carbon, and sulfate. The first two aerosol types are provided in three radius size bins: 0.03-0.5, 0.5-5 and 5-20 μm and 0.03–0.55, 0.55–0.9 and 0.9–20 μm , respectively. The different IFS aerosol types are treated as externally mixed (Inness et al., 2019a).

Here we use IFS day-1 ~~forecasts~~ (referred here as IFS) forecasts (initiated at 00:00Z) of temperature, mean sea level pressure (mslp), omega vertical velocity, divergence, u and v wind components, ~~PM₁₀ and PM_{2.5} surface concentrations,~~ mass mixing ratio of 11 aerosol variables, total AOD at 550 nm (AOD₅₅₀ or just AOD), sea salt AOD, dust AOD, organic matter AOD, black carbon AOD, sulfate AOD and carbon monoxide (CO) mass mixing ratio. ~~The~~ Also used are day-1 to day-3 forecast data of PM₁₀ and PM_{2.5} concentrations are calculated as follows surface concentrations, which are derived according to the following formula:

$$PM_{10} = \rho \cdot (SS1 / 4.3 + SS2 / 4.3 + DD1 + DD2 + 0.4 \cdot DD3 + OM1 + OM2 + SU1 + BC1 + BC2) \quad (1)$$

$$PM_{2.5} = \rho \cdot (SS1 / 4.3 + 0.5 \cdot SS2 / 4.3 + DD1 + DD2 + 0.7 \cdot OM1 + 0.7 \cdot OM2 + 0.7 \cdot SU1 + BC1 + BC2) \quad (2)$$

where ρ the air density; SS1 the ~~sea salt~~ sea salt radius size bin 1 (0.03-0.5 μm), SS2 the ~~sea salt~~ sea salt radius size bin 2 (0.5-5 μm), DD1 the desert dust radius size bin 1 (0.03–0.55 μm), DD2 the desert dust radius size bin 2 (0.55–0.9 μm), DD3 the desert dust radius size bin 3 (0.9–20 μm), OM1 the ~~organic matter bin 1~~ hydrophobic organic matter, OM2 the ~~organic matter bin 2~~, SU1 the sulphur bin hydrophilic organic matter, BC1 the ~~black carbon bin 1 and~~ hydrophobic black carbon, BC2 ~~black carbon bin 2~~ (, last access: 18 February 2020) the hydrophilic black carbon and SU1 the aerosol sulfate (ECMWF, 2020). A detailed description and evaluation of the aerosol scheme used in IFS can be found in Rémy et al. (2019). To unravel the impact of chemical data assimilation on aerosol burden representation during the examined event, an additional IFS control run without data assimilation (no DA) is also used for intercomparison.

2.2 CAMS regional forecast system

The CAMS regional forecast system provides the ensemble median (RegEns) and members of the European-scale air quality forecasts for every hour up to four days in advance. The products have a 0.1° horizontal resolution and are based on state-of-the-art numerical air quality models developed in Europe: CHIMERE from INERIS (National Institute for Industrial Environment

and Risks) (Menut et al., 2014), DEHM from Aarhus University (Christensen, 1997), EMEP from MET-Norway (Simpson et al., 2012), EURAD-IM from the University of Cologne (Memmesheimer et al., 2004), GEM-AQ from IEP-NRI (Institute of Environmental Protection, Polism Ministry of Environment) (Kaminski et al., 2008), LOTOS-EUROS from KNMI (Royal Netherlands Meteorological Institute) and TNO (Netherlands Organisation for Applied Scientific Research) (Manders et al., 5 2017), MATCH from SMHI (Swedish Meteorological and Hydrological Institute) (Robertson et al., 1999), MOCAGE from Météo-France (Guth et al., 2016) and SILAM from FMI (Finnish Meteorological Institute) (Sofiev et al., 2015). More details on the regional air quality systems can be found at [CAMS \(2020c\)](#). The global ECMWF IFS forecasts provide the meteorological and the chemical boundary forcing to the regional model suite and CAMS provides the emissions. [Several CAMS regional models assimilate PM₁₀ and PM_{2.5} surface observations from several various stations of the EEA's \(European Environment Agency\) Air Quality e-reporting database, but not satellite aerosol products. More specifically, during the period of interest \(October 2017\), CHIMERE and EURAD were assimilating both PM₁₀ and PM_{2.5}, MOCAGE only PM₁₀ and, finally, SILAM and MATCH were assimilating only PM_{2.5}. PM₁₀ and PM_{2.5} concentrations in the regional models are calculated using simulated aerosol fields specific to each regional model. All models are validated operationally \$\ominus\$ \(CAMS, 2020b\) and a posteriori \$\ominus\$ \(CAMS, 2020a\), and operational verification results are available daily for six chemical species \(ozone, nitro- 10 gen dioxide, sulfur dioxide, carbon monoxide, PM₁₀ and PM_{2.5}\). It has to be noted that for the examined period \(October 2017\) DEHM and GEM-AQ models are not included in \[RegENSRegEns\]\(#\), as they were recently added in the CAMS regional forecast system. RegEns day-1 \(referred here as RegEns\), day-2 and day-3 forecast data \(initiated at 00:00Z\) of PM₁₀ and PM_{2.5} surface concentrations are used, to analyze and evaluate the performance of CAMS regional forecast system during the examined event.](#)

20 2.3 Validation data

To evaluate the CAMS forecast systems ability to capture the spatial and temporal distribution of aerosols in the atmosphere, AOD₅₅₀ data from the two MODIS sensors aboard EOS Terra (equator crossing time 10:30 LT ([Local Time](#))) and Aqua (equator crossing time 13:30 LT) satellites are used along with aerosol subtype data from CALIOP/CALIPSO (equator crossing time \sim 13:30 LT). MODIS is a 36-band imaging radiometer with a viewing swath of 2330 km offering al- 25 most daily global coverage (Salomonson et al., 1989). In this work, level-2 data with a resolution of 10 x 10 km² at nadir from the MODIS Collection 6.1 combined Dark Target algorithm (DT) and Deep Blue algorithm (DB) scientific dataset AOD_550_Dark_Target_Deep_Blue_Combined are processed. There are two different DT algorithms, one for retrievals over land (Kaufman et al., 1997; Remer et al., 2005; Levy et al., 2013) and one for water surfaces (Tanré et al., 1997; Remer et al., 2005; Levy et al., 2013), while DB currently delivers retrievals over all land types (Hsu et al., 2013; Sayer et al., 2013, 30 2014, 2015) despite the fact that originally was developed for bright land surfaces only (Hsu et al., 2004). For the quantitative evaluation of the CAMS forecasts the MODIS/Terra and Aqua data were merged and brought to the CAMS native grid.

CALIOP is a spaceborne lidar instrument (Hunt et al., 2009) providing profiles of aerosol and cloud-related properties (Winker et al., 2009) within the first 30 km of the atmosphere by measuring the backscatter signals and the polarization of the backscattered light. The CALIPSO algorithm identifies distinct atmospheric layers (clean air, aerosols, clouds, surface, etc.)

(Vaughan et al., 2009; Kim et al., 2018) and attributes a specific aerosol subtype (marine, dust, polluted continental/smoke, clean continental, polluted dust, elevated smoke, dusty marine, polar stratospheric cloud aerosols, volcanic ash, sulfate/other) to each of them (Omar et al., 2009). In this work, data from the CALIOP/CALIPSO version 4.20 level-2 product (Kim et al., 2018) with a horizontal resolution of 5 km and a vertical resolution of 30 m (for heights below ~20 km) are processed.

- 5 To assess CAMS forecast systems' performance in reproducing the impacts on air quality during the Ophelia passage ground-based observations are used. Measurements of PM₁₀ and PM_{2.5} surface concentrations from 8 rural background stations (see Table 1 for details) are obtained from the EMEP (Tørseth et al., 2012) network through the EBAS database [\(EBAS, 2020\)](#) for the time period from 10 October 2017 to 20 October 2017. The stations are located over ~~the broader western European areas where~~ [western Europe and away from the](#) dust and biomass burning ~~transport occurred~~ [sources, lie across the plumes of](#) [high AOD loadings, exhibiting significant increases in PM₁₀ and PM_{2.5} surface concentrations during the examined event.](#)
- 10 Data are provided with 1-hour temporal resolution, yet a 3-hour resolution is used for direct comparison with the IFS data. It has to be noted that from the examined stations only GB0043R and GB0048R for PM₁₀, and GB0048R for PM_{2.5}, are listed as assimilation stations for the CAMS regional models.

2.4 [Statistical metrics](#)

- 15 [To evaluate the performance of CAMS forecast systems with respect to observational data, the following statistical metrics are used.](#)

- a) [The temporal correlation of CAMS models with observations is assessed with the Pearson correlation coefficient \(R\) that measures the strength of their linear association, ranging between +1 and -1:](#)

$$R = \frac{N \sum M_i O_i - \sum M_i \sum O_i}{\sqrt{N \sum M_i^2 - (\sum M_i)^2} \sqrt{N \sum O_i^2 - (\sum O_i)^2}} \quad (3)$$

- 20 [where \$M_i\$ and \$O_i\$ are the modeled and observed values, respectively, and N is the number of the sample.](#)

- b) [The model error is estimated using the fractional gross error \(FGE\) which ranges between 0 and 2, and behaves symmetrically with respect to under- and overestimation:](#)

$$FGE = \frac{2}{N} \sum_i^N \left| \frac{M_i - O_i}{M_i + O_i} \right| \quad (4)$$

- [where \$M_i\$ and \$O_i\$ are the modeled and observed values, respectively, and N is the number of the sample.](#)

3 Results

3.1 Storm Ophelia and transport pathways

Hurricane Ophelia occurring in October 2017 was an exceptional low pressure system, as it had unique characteristics as an Atlantic hurricane, it caused the death of three people and extended damages during its passage over Ireland (BBC, 2017), and indirectly affected the air quality and the atmospheric composition over several western European areas. Ophelia initiated as a non-tropical low pressure system over the Atlantic at the southwest of the Azores in early October 2017, and didn't have the fate as that of a common Atlantic tropical storm that it heads to the west (Stewart, 2018). Instead, Ophelia marched northeastwards initially reaching coastal Portugal, and subsequently losing its tropical characteristics downgrading to an extratropical cyclone (ex-hurricane) it followed a northern path towards the Great Britain, becoming the easternmost major Atlantic hurricane ever recorded (Stewart, 2018). The aforementioned route of Ophelia resulted from the guidance of an upper level trough located over the Atlantic (Stewart, 2018; Rantanen et al., 2020). Figure 1 presents the prevailing synoptic conditions in the middle troposphere (500 hPa), as seen from IFS, during the period from 12Z on 14 October 2017 to 00Z on 16 October 2017. A mid-latitude trough dominates over the central Atlantic, with the surface low pressure system located ahead of the trough and to the east of it, being driven from the northeasterly oriented wind flow (Fig. 1a and b). On 12Z of October 15th and as the trough axis turns negatively tilted the flow becomes mostly southerly (Fig. 1c) dragging Ophelia towards the Ireland (Fig. 1d). Although after 12Z of October 14th Ophelia moved over sea surface temperatures (SSTs) of about 25°C which in general do not support hurricane intensification (Stewart, 2018), the alternative fuel for Ophelia to maintain its strength was found in the enhanced upper level divergence on the eastern flank of the trough (not shown). This resulted in convection reinforcement, as revealed from the enhanced upward vertical velocity values at 500 hPa exceeding 2 Pa/s (Fig. 1).

From 00Z of October 13th and onwards, Saharan dust being transported through the trade winds over the west coast of Africa, is exposed to the southerlies on the east side of Ophelia system, setting up a dust outbreak that gradually moves to the north. On 12Z of October 14th a plume of high IFS dust mass mixing ratio exceeding 80 µg/kg is found west of the Iberian Peninsula up to approximately 700 hPa, as depicted in Figs. 2a and b. During the next 24 hours and as the surface low pressure system is traveling further north (at ~ 40°N on 12Z of October 15th), the dust particles entrained from the meridional flow at the east of Ophelia's periphery form an extended plume of high dust loadings reaching the southeast coast of UK (Figs. 2c, d, e and f). Throughout the following 12 hours the plume expands to the north and east passing over northwestern France and southern UK (Figs. 2g and h). On 00Z of October 16th the Ophelia storm merges with the frontal system as can be seen in Fig. 2g and Fig. 3a, while smoke from wildfires that have been burning across the Iberian Peninsula is evident at 850 hPa, as high CO volume mixing ratio (> 200 ppbv), uplifted through the Warm Conveyor Belt (Osborne et al., 2019) on the southeast part of Ophelia where upward motion dominates (Fig. 2g and Fig.S1 in the Supplement). From this moment and until 18Z of October 16th, dust and smoke particles gather on the warm and cold front of Ophelia's warm sector, respectively, transported over western Europe through the frontal system

(Figs. 3c, e, and g). This distinct discretization of dust and biomass burning within the warm sector of Ophelia is well illustrated by the 3-D fields of dust and CO mixing ratio exceeding $80 \mu\text{g}/\text{kg}$ and 200 ppbv respectively, resembling the shape of the warm and cold front (Figs. 3d, f, and h). Noteworthy is the uplift of smoke over northwest Iberian Peninsula from 18Z on 16 October to 06Z on 17 October (Figs. 2l), due to the profound upward motions induced from strong lower convergence and upper divergence (Fig.S1 in the Supplement) at the eastern flank of the upper level trough (not show). This elevated smoke plume is drifted during the next 24 hours from the northeasterly flow towards UK (Figs. 2n and p). The Ophelia storm starts dissipating from 00Z of October 17th onwards fading away over Norway on 00Z of October 18th. A graphical representation of the IFS dust and CO transport along with the synoptic evolution during the passage of Ophelia (from 00Z of October 12th 2017 to 21Z of October 20th 2017) is provided as animation in the Supplement.

3.2 Aerosol atmospheric composition

Here we explore the impact of Ophelia's passage on the aerosol atmospheric composition over the broader European region, using CAMS global forecast data and satellite observations. The MODIS/Terra and Aqua AOD₅₅₀ spatial distribution for the time period from 14 October 2017 to 18 October 2017 is presented in Figure 4 (left column). It should be noted that these data don't give an independent validation as they are also assimilated in the IFS. High aerosol loadings (> 0.4) are observed at the west of the Iberian Peninsula at 14 October (Fig. 4a), while one day later the aerosol plume is extended further north and east over France, English Channel and Celtic Sea (Fig. 4d). On 16 and 17 October, high AOD₅₅₀ values are seen over North and Baltic Sea, France, Belgium, the Netherlands and Germany (Fig. 4g and j), while on 18 October the plume is found over Germany and Poland (Fig. 4m). The observed buildup of high AOD loadings over Europe is in line with the aforementioned description of the Ophelia storm and the associated transport pathways. To evaluate the CAMS global AOD₅₅₀ forecast product during this period, the respective AOD₅₅₀ fields are also shown in Fig. 4 (middle column). Overall, there is a good agreement between the CAMS global forecast product and the satellite observations, at least in qualitative terms, as the forecasted high AOD loadings resemble that of observations. To assess the impact of AOD data assimilation on IFS AOD forecast, the differences of AOD₅₅₀ with and without the use of data assimilation are also shown in Fig. 4 (right column), revealing that data assimilation boosts AOD values near the examined AOD plumes, while near the dust sources over Africa it mostly suppresses them. In addition, to quantitatively validate the CAMS global AOD forecast, a comparison of spatially and temporally collocated AOD₅₅₀ data between IFS (with and without the use of data assimilation) and MODIS/Terra and Aqua is performed for two box regions; the whole domain appearing in Fig. 4 and a smaller domain in northwestern Europe appearing in Fig. 9d. As depicted from the scatter plots of Figs. 5a and b for the large domain, the use of data assimilation improves the CAMS global forecast performance increasing the correlation (from 0.72 to 0.77) and reducing the ~~bias (fractional gross error (fge))~~ error (FGE) from 0.59 to 0.4) with respect to the control simulation (no DA). For a more "event-based" evaluation of CAMS global AOD forecast, the respective scatter plots for a small domain in northwestern Europe where the AOD plume is found are shown in Figs. 5c and d, indicating less agreement with the satellite observations and a slight improvement due to data

assimilation implementation (correlation increase from 0.54 to 0.56, and [fge-FGE](#) decreases from 0.48 to 0.44). Overall, the CAMS global AOD forecast product tends to overestimate for low [observed](#) AOD values and vice versa, a behavior that was also observed in a previous version of the ECMWF global atmospheric composition reanalysis dataset (MACC (Monitoring Atmospheric Composition and Climate); see Georgoulias et al. (2018) for details).

5 To identify the dominant IFS aerosol types within the high AOD plume, the [modeled](#) percentage (%) contribution of each aerosol type to the total AOD, as shown in Fig. 4, is presented in Fig. 6 for the same period. On 12Z of October 14th, the dominant source of the AOD loading found at the west of the Iberian Peninsula is mineral dust by 60-70%, while the dust contribution over UK is up to 20%, as depicted from Fig. 6a. One day later and as the AOD plume moves to the north and east, dust continues to dominate with percentage contribution of up to 45% over UK (Fig. 6f). On 10 12Z of October 16th, the dust AOD contribution over UK ranges with a percentage between 20-35%, while over the North Sea and the Netherlands it reaches values up to 65% (Fig. 6k). The biomass burning emission trails originating from northwest Iberian Peninsula are also a significant input for AOD, with black carbon and organic matter aerosols contributing up to 20% and 40% over UK, respectively (Figs. 6m and n). [Such black carbon contributions are considered high, being similar to climatological contributions over global fire hot spots \(e.g. summertime central southern Africa; Penning de Vries et al. \(2015\)\).](#) 15 The elongated plume of high AOD values on 12Z of October 17th as seen from both the satellite observations and the CAMS global forecast (Figs. 4j and k), consists of up to ~50% of organic matter aerosols (Fig. 6s), up to ~25% of black carbon aerosols (Fig. 6r) and up to ~20% of dust aerosols (Fig. 6p), while over the Baltic States the major contributor is mineral dust with ~50%. This is consistent with the transport pathways of dust and biomass burning aerosols shown in Fig. 2, indicating that fire-originated aerosols dominate over northern 20 coastal Europe, while dust aerosols remnants from Ophelia's warm front passage prevail over the Baltic States. A similar situation also occurs one day later as depicted from Figs. 6u, w and x. It is worth noting, that during the examined period sea salt aerosols are dominating over the Atlantic Sea, following the evolution of Storm Ophelia, as they are produced and transported due to the intense winds near Ophelia (Figs. 6b, g, l, q, v).

To assess the performance of CAMS global forecast in representing the vertical distribution of aerosol types during 25 the passage of Ophelia, CALIPSO aerosol subtype satellite data were also utilized. Two CALIPSO tracks that crossed the aerosol plume during the examined period were selected for 15/10/2017 and 17/10/2017 with an overpass time around 13:00Z and 12:45Z, respectively (tracks are shown on the top left of Figs. 7a and c). For the selected CALIPSO tracks, spatially (horizontally and vertically) and temporally (at 12Z) collocated IFS aerosol mass mixing ratio data were extracted. Subsequently, the dominant (in terms of mass mixing ratio) aerosol type was identified, namely, sea 30 salt, dust, carbonaceous aerosols (organic matter and black carbon) and sulfates, [following Georgoulias et al. \(2020\)](#). A dominant aerosol type was set only when the highest mass mixing ratio from the four subtypes exceeded 1 $\mu\text{g}/\text{kg}$. Figure 7 presents the vertical cross sections of CALIOP/CALIPSO aerosol subtypes and IFS dominant aerosol type along the two selected CALIPSO tracks. On 12Z of October 15th, a distinct presence of dust and polluted dust aerosols is observed between 44°N and 57°N, with polluted dust being confined within the first ~2 km. Moreover, polluted

continental/smoke aerosols are also detected over the UK (>3650°N) up to approximately 1 km. Further south and over the Mediterranean Sea (36°N-43°N), sea salt and dusty marine aerosols are mostly dominating indicating a dust cross over the ~~mediterranean~~Mediterranean, while southern than 36°N, over North Africa, polluted dust is observed (Fig. 7a). The respective vertical cross section of IFS dominant aerosol type indicates the dominance of dust over the latitudinal bands of 40°N-55°N and 30°N-35°N, and of carbonaceous aerosols near the surface between ~~4943°N~~4943°N and 55°N, while ~~a mixture of sea salt and carbonaceous aerosols is~~ primarily carbonaceous aerosols and secondarily sea salt aerosols are identified over the Mediterranean Sea (Fig. 7b), being in a satisfactory agreement with the CALIOP/CALIPSO observations. For the second selected CALIPSO track on 12Z of October 17th, dust and polluted dust are detected at ~~45°N-50N-52°N~~45°N-50N-52°N and 30°N-35°N up to 3 km, with dust also observed in the upper troposphere (~10 km) over France, while elevated smoke is identified over the North Sea at ~4 km (Fig. 7c). The latitudinal bands of IFS dust ~~aerosols dominance are~~ dominance is consistent with CALIPSO, ~~yet, as well as the~~ fire-originated aerosols (carbonaceous aerosols) which are found over the North Sea ~~higher (above 5 above 2 km) compared to CALIPSO~~ (Fig. 7d). ~~This might be due to sea salt aerosol overestimation that prevents carbonaceous aerosols to appear as the dominant type below 5~~ Yet, over the Mediterranean carbonaceous aerosols prevail in IFS, while marine and dusty marine aerosols in CALIOP/CALIPSO.

3.3 Air quality

The implications of dust and biomass burning transport during the Ophelias's passage for European air quality are hereafter discussed. Figure 8 illustrates the CAMS global and regional PM₁₀ and PM_{2.5} surface concentrations over Europe for the time period from 18Z on 15 October 2017 to 06Z on 17 October 2017. Overall, the spatial patterns of particulate matter concentrations in the CAMS products agree well, reproducing the plume of high aerosol loadings. Yet, there are some differences between the global and regional forecasts which are as follows: a) RegEns exhibits lower concentrations compared to IFS for both coarse and fine particulate matter, b) IFS has higher concentrations near the center of Ophelia probably due to higher sea salt contribution, c) the enhanced PM₁₀ and PM_{2.5} concentrations over Portugal in IFS due to assimilated fire emissions (GFAS) are not seen in RegEns, d) RegEns exhibits higher concentrations in both PM₁₀ and PM_{2.5} over the Black Sea and the Baltic States compared to IFS. The aforementioned inconsistencies are likely due to the different definition of PM₁₀ and PM_{2.5} in IFS and each CAMS regional air quality model, the different aerosol schemes and horizontal resolution used in CAMS global and regional models, and the different assimilation approach in the CAMS regional air quality models relative to IFS.

To evaluate the CAMS forecast systems and the role of data assimilation in reproducing the enhancements in surface PM₁₀ and PM_{2.5} concentrations, we employ ground-based measurements from the EMEP network from 8 rural stations (see Table 1 for further info and Fig. 9d for stations location) for intercomparison. For brevity, only the results from three EMEP stations are presented (Guipry FR0024R, France; Chilbolton Observatory GB1055R, Great Britain; Cabauw Wielesekade NL0644R, the Netherlands), while results for the rest of the stations are provided in the Supplement. Figure 9

presents the PM₁₀ surface concentrations from observations, IFS ([day-1 to day-3 forecasts](#)), IFS without data assimilation and RegEns ([day-1 to day-3 forecasts](#)), along with the percentage (%) contribution of each aerosol type to IFS PM₁₀ surface concentrations, during the time period from 10 October 2017 to 20 October 2017. At Guipry (France), after 12Z of October 14th and up to October 16th an increase in PM₁₀ surface concentrations is observed, also seen in IFS and RegEns; however the early peak in observations is not captured by CAMS forecast systems (Fig. 9a, top). The percentage contribution of each IFS aerosol type to IFS PM₁₀ concentrations indicates that the enhancement in PM₁₀ surface levels is initially (00Z on 15 October) due to organic matter aerosols (~75%), probably related to transported smoke from fires burning over the Iberian Peninsula (visual inspection of MODIS Fires and Thermal Anomalies product, source: [NASA \(2020\)](#)), while afterwards (21Z on 15 October) the transport of dust induces a significant contribution from dust aerosols (~53%) as well (Fig. 9a, bottom). On 12Z of October 16th a sharp peak is depicted in observed PM₁₀ concentrations reproduced from both IFS and RegEns, yet overestimated from IFS, which seems to result from sea salt transport from the west and on the south of Ophelia (see flow at 850 hPa at Fig. 2i). IFS exhibits an increase of PM₁₀ levels on 12Z of October 18th due to dust and organic matter aerosols which is not seen in RegEns and observations. Similar performance for CAMS forecast systems is obtained for PM_{2.5} over Guipry, except that the peak on 18 October in IFS is also confirmed from observations but to a lower extent (Fig. 10a). Further north and over Chilbolton Observatory (Great Britain), after 15Z of October 15th the observed PM₁₀ surface concentrations increase up to 38 µg/m³, with IFS peaking with a 6 hours delay (Fig. 9b, top) mostly due to dust and organic matter aerosols (Fig. 9b, bottom). The secondary peak seen in observations on 12Z of October 16th is synchronously captured by RegEns and IFS, with IFS clearly overestimating it. A second wave of dust and biomass burning aerosols transport is affecting PM₁₀ surface concentrations over the site on 15Z of October 18th as revealed from IFS which is in agreement with observations but not seen in RegEns. Regarding PM_{2.5}, there is a similar level of agreement with that of PM₁₀ between the CAMS forecast systems and the observations (Fig. 10b). Finally, over Cabauw Wielekade (the Netherlands), the observed PM₁₀ surface concentrations exhibit a more noisy structure, with CAMS forecast systems reproducing only the general pattern of variability, missing the observed peaks on 15Z of October 17th and 18th (Fig. 9c). Similarly for PM_{2.5}, where the observed peaks are more clear, IFS and RegEns capture the big picture of PM_{2.5} increases (Fig. 10c). Results for the rest five EMEP stations are provided in Figs. S2 and S3 in the Supplement. Overall, RegEns exhibits a better performance in terms of temporal variability and bias compared to IFS. As depicted from the scatter plot in Fig. 11a the correlation coefficient of RegEns with observations is higher compared to that of IFS with observations (red points mostly lie above the dashed line of Fig. 11a), while the f_{ge-FGE} values for RegEns are lower with respect to that of IFS (red points lie below the dashed line of Fig. 11b). Concerning the impact of data assimilation on IFS forecast of near surface particulate matter, as it is depicted from Fig. 11 there is no clear improvement neither in bias nor in the representation of the temporal variability. It is likely that the assimilation of total-column AOD does not have a strong impact on surface-layer concentrations, especially in regions with significant surface fluxes.

[The capability of IFS and RegEns systems to forecast the observed PM₁₀ and PM_{2.5} surface concentrations two and three days in advance, is finally discussed. As depicted in Figure 9, IFS day-2 and day-3 forecasts reproduce the distinct](#)

increases in observed PM_{10} surface concentrations exhibiting similar FGE values but lower correlation scores (in most of the stations) compared to day-1 forecast (Fig. 11c and d). The same applies in the case of $PM_{2.5}$ (Fig. 10), except that the correlation scores for IFS day-2 and day-3 forecasts are not systematically lower than that of day-1 forecast (Fig. 11c and d). As regards the RegEns, although it fairly predicts the observed peaks in PM_{10} and $PM_{2.5}$ up to three days in advance (Fig. 9 and 10), there is a systematic deterioration of its performance in terms of temporal variability over forecast time. More specifically, the correlation coefficient decreases from day-1 to day-2 forecast and from day-2 to day-3 forecast for almost all examined stations (Fig. 11e).

4 Conclusions

The main objectives of this work were to analyze a complex case study of aerosol transport over Europe driven by Storm Ophelia in mid-October 2017 in CAMS forecast systems, and assess their performance with respect to aerosol atmospheric composition and air quality. To this end, CAMS day-1 forecast data were used along with satellite and ground-based observations. The most notable findings of this work can be summarized as follows:

- African dust aerosols were transported towards northwestern Europe on the east side of Storm Ophelia forming an extended dust plume reaching the Baltic States. On its passage, Ophelia lifted and drifted smoke aerosols from fires burning over the Iberian Peninsula carrying them over several European areas, such as France, Great Britain and the Netherlands. After 00Z of October 16th, dust/smoke aerosols are found on the warm/cold front of Ophelia being transported within its warm sector.
- The dependent evaluation against MODIS satellite observations reveals a satisfactory agreement of CAMS global AOD_{550} is in satisfactory agreement with the MODIS satellite observations ($r(R)=0.77$ and $fgeFGE=0.4$), while the comparison against the IFS no DA simulation indicates an improvement of IFS performance in forecasting AOD_{550} ($r(R)$ increases from 0.72 to 0.77, and $fgeFGE$ decreases from 0.59 to 0.4), due to the implementation application of data assimilation.
- The speciation of IFS AOD_{550} fields indicates that African dust, organic matter from Iberian biomass burning and sea salt sprayed in the vicinity of Ophelia, are the major aerosol contributors to the plume of high AOD_{550} .
- The CALIOP/CALIPSO and the spatially and temporally collocated IFS cross sections of dominant aerosol types, reveal a good qualitative performance of IFS, as it generally manages to reproduce the type and location of aerosols during the passage of Ophelia.
- In terms of air quality, comparison with ground-based measurements from EMEP stations reveals that both IFS and RegEns are able to reproduce the observed signal of increase in PM_{10} and $PM_{2.5}$ surface concentrations, yet exhibiting several inconsistencies on the quantitative and temporal representation of the observed peaks.

- [The For the examined event, the](#) CAMS regional system seems to better predict the 3-h PM_{10} and $PM_{2.5}$ surface concentrations compared to CAMS global system, with an increase/decrease of the correlation coefficient/~~fge~~-[FGE](#) values in all examined stations for PM_{10} (except Cabauw Wielsekade) and $PM_{2.5}$, respectively.
- As regards the role of data assimilation, even though it improves the CAMS global forecast performance for AOD there is no such indication for particulate matter at near surface.
- [A deterioration of the RegEns forecast performance is found over forecast time for both \$PM_{10}\$ and \$PM_{2.5}\$, characterized by a decrease of the correlation coefficient for the vast majority of the examined stations, which is partially seen in IFS for the case of \$PM_{10}\$.](#)

To summarize, the current analysis and evaluation study highlights that CAMS global forecast system is able to forecast the observed aerosol loadings over Europe induced from the transport of dust, biomass burning and sea salt aerosols through Ophelia's Storm passage in mid-October 2017. Forecasting coarse and fine particulate matter surface concentrations during Ophelia turns out to be more challenging, yet the CAMS global and regional systems exhibit a reasonable performance, clearly open to further improvement and development in the direction of supporting social, economic and health policies.

Author contributions. DA designed the study, performed the analysis and wrote the paper. AKG provided the MODIS/Terra and Aqua satellite data, the CALIOP/CALIPSO satellite data and contributed in the analysis and writing of the paper. All authors contributed in reviewing and editing of the manuscript, and interpretation of the results. JF provided the IFS control simulation without the use of data assimilation. HE is coordinating CAMS84 (Global and regional a posteriori evaluation and quality assurance). EK is the Principal Investigator and coordinates the contribution of Aristotle University of Thessaloniki in CAMS84.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1. Observational data used in the present study.

EMEP ground-based stations					
Name	Country	Code	Location	PM ₁₀	PM _{2.5}
La Coulonche	France	FR0018R	0.45° W,48.63° N	yes	no
Guipry	France	FR0024R	1.84° W,47.83° N	yes	yes
Narberth	Great Britain	GB0043R	4.69° W,51.78° N	yes	no
Auchencorth	Great Britain	GB0048R	3.24° W,55.79° N	yes	no
Moss					
Chilbolton Obser- vatory	Great Britain	GB1055R	1.44° W,51.15° N	yes	yes
Kollumerwaard	the Netherlands	NL0009R	6.28° E,53.33° N	yes	yes
De Zilk	the Netherlands	NL0091R	4.5° E,52.3° N	yes	yes
Cabauw	the Netherlands	NL0644R	4.92° E,51.97° N	yes	yes
Wielsekade					
Satellite observations					
Sensor/Satellite	Type	Overpass	Coverage	Resolution	Product
MODIS/Terra	passive	10:30 LT	daily	10 x 10 km ²	AOD ₅₅₀
MODIS/Aqua	passive	13:30 LT	daily	10 x 10 km ²	AOD ₅₅₀
CALIOP/CALIPSO	active	~ 10:30 LT	16-day repeat cycle	hor.: 5 km, ver.: 60 m (<20 km)	aerosol subtype

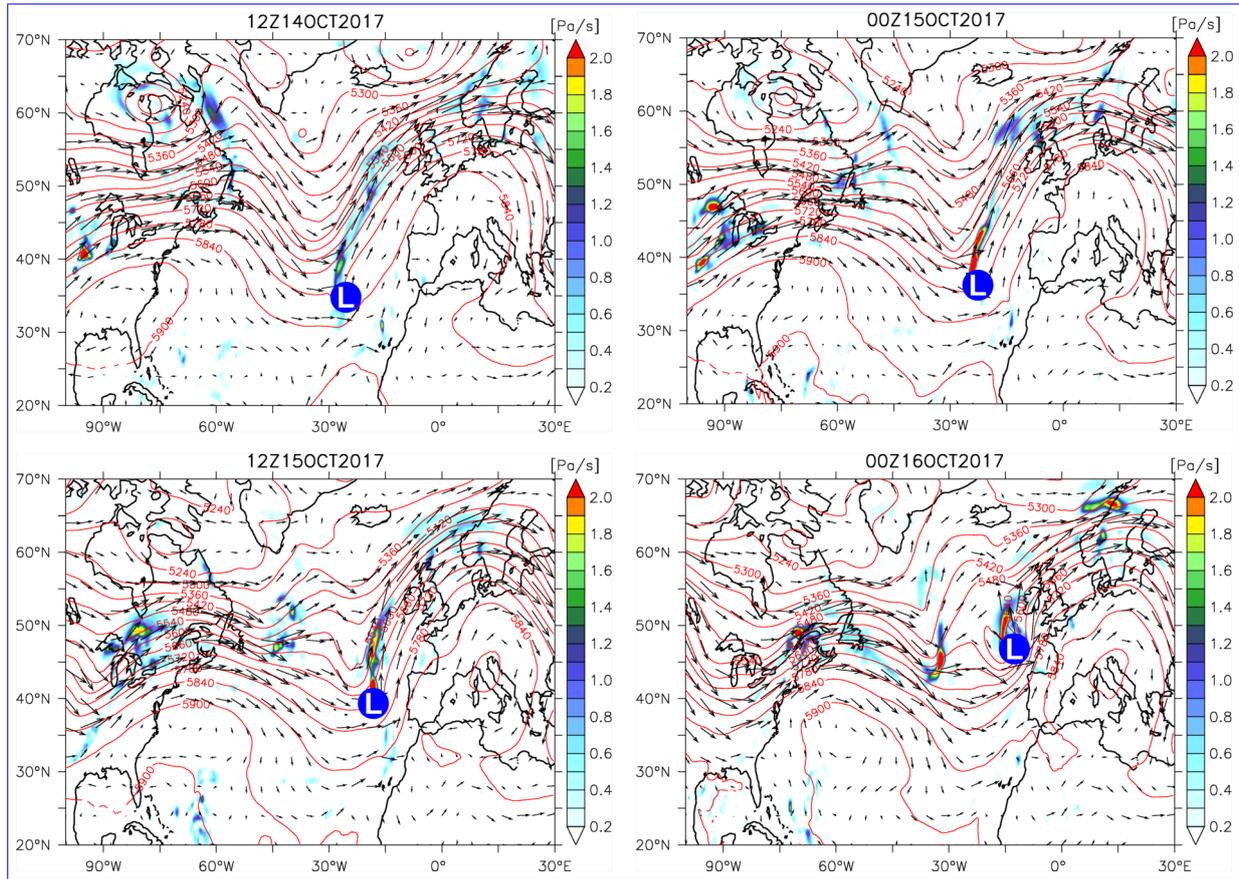


Figure 1. IFS geopotential height (in gpm; red contours), negative of omega vertical velocity ($-dp/dt$) (in Pa/s, color shaded) and wind direction (black vectors) at 500 hPa, during the period from 12:00Z on 14 October 2017 to 00:00Z on 16 October 2017 (12 h interval) (a, b, c, d). Also shown is the location of the surface low pressure system denoted with the capital letter L.

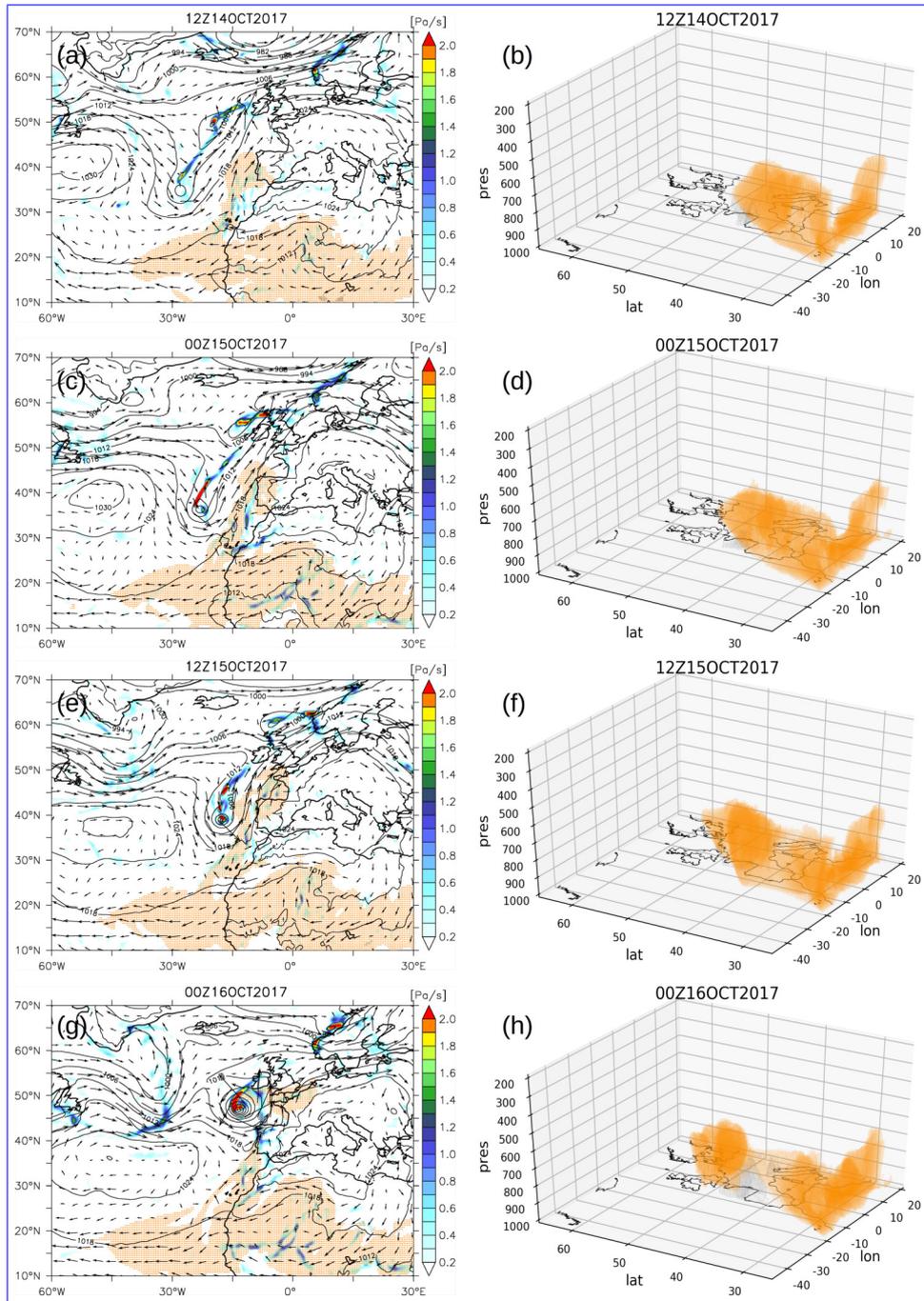


Figure 2. First part of the figure

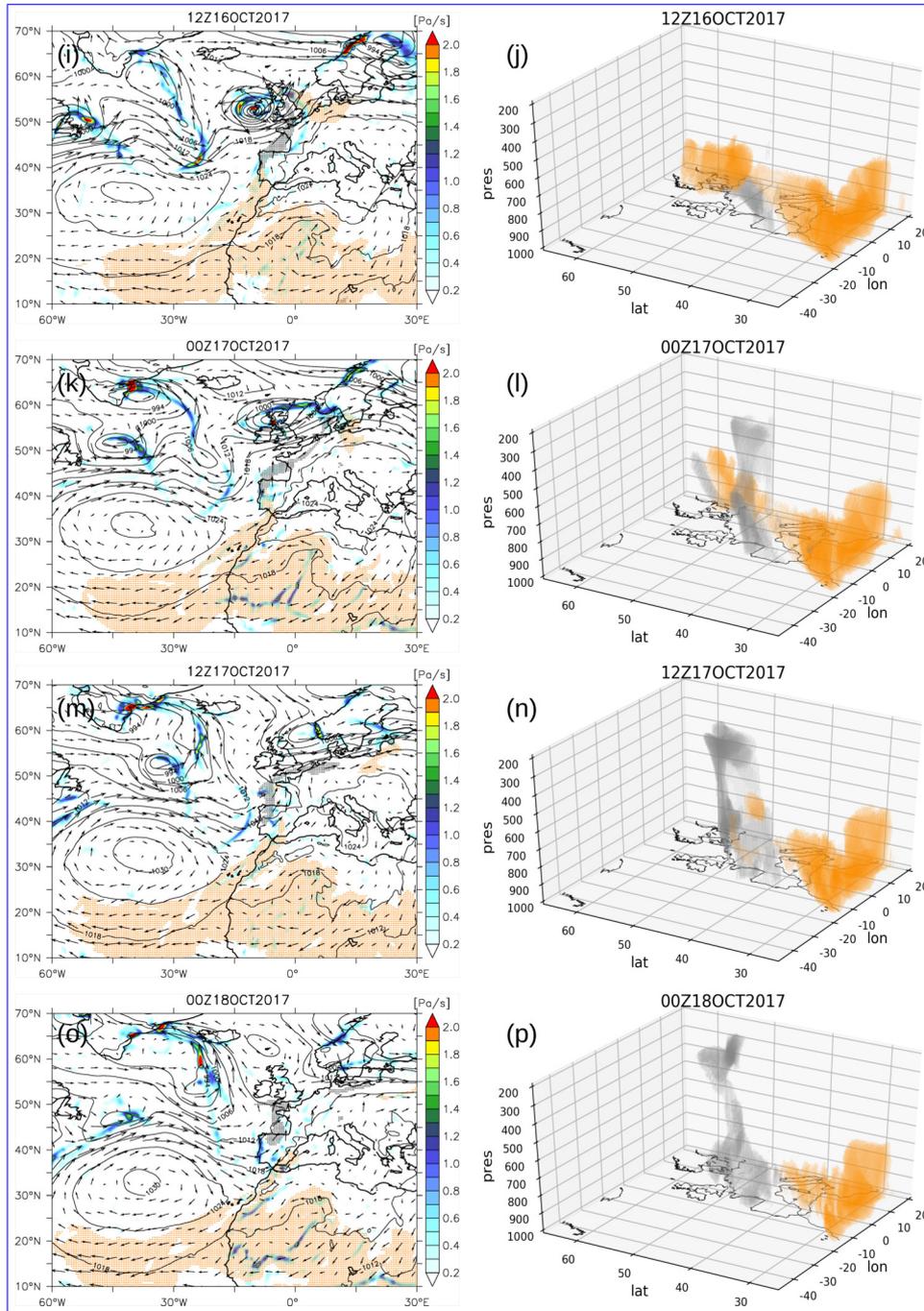


Figure 2 (Continued). IFS mslp (in hPa; black contours), negative of omega vertical velocity ($-\text{dp}/\text{dt}$) (in Pa/s , color shaded), wind direction (black vectors), dust with mass mixing ratio exceeding $80 \mu\text{g}/\text{kg}$ (orange shaded) and CO with volume mixing ratio exceeding 200 ppbv (grey shaded) at 850 hPa during the period from 12:00Z on 14 October 2017 to 00:00Z on 18 October 2017 (12 h interval) (Figures 2a, 2c, 2e, 2g, 2i, 2k, 2m, 2o). Three-dimensional (longitude, latitude, pressure (hPa)) spatial distribution of IFS dust with mass mixing ratio exceeding $80 \mu\text{g}/\text{kg}$ (orange shaded) and IFS carbon monoxide (CO) with volume mixing ratio exceeding 200 ppbv (grey shaded) during the same period (Figures 2b, 2d, 2f, 2h, 2j, 2l, 2n, 2p).

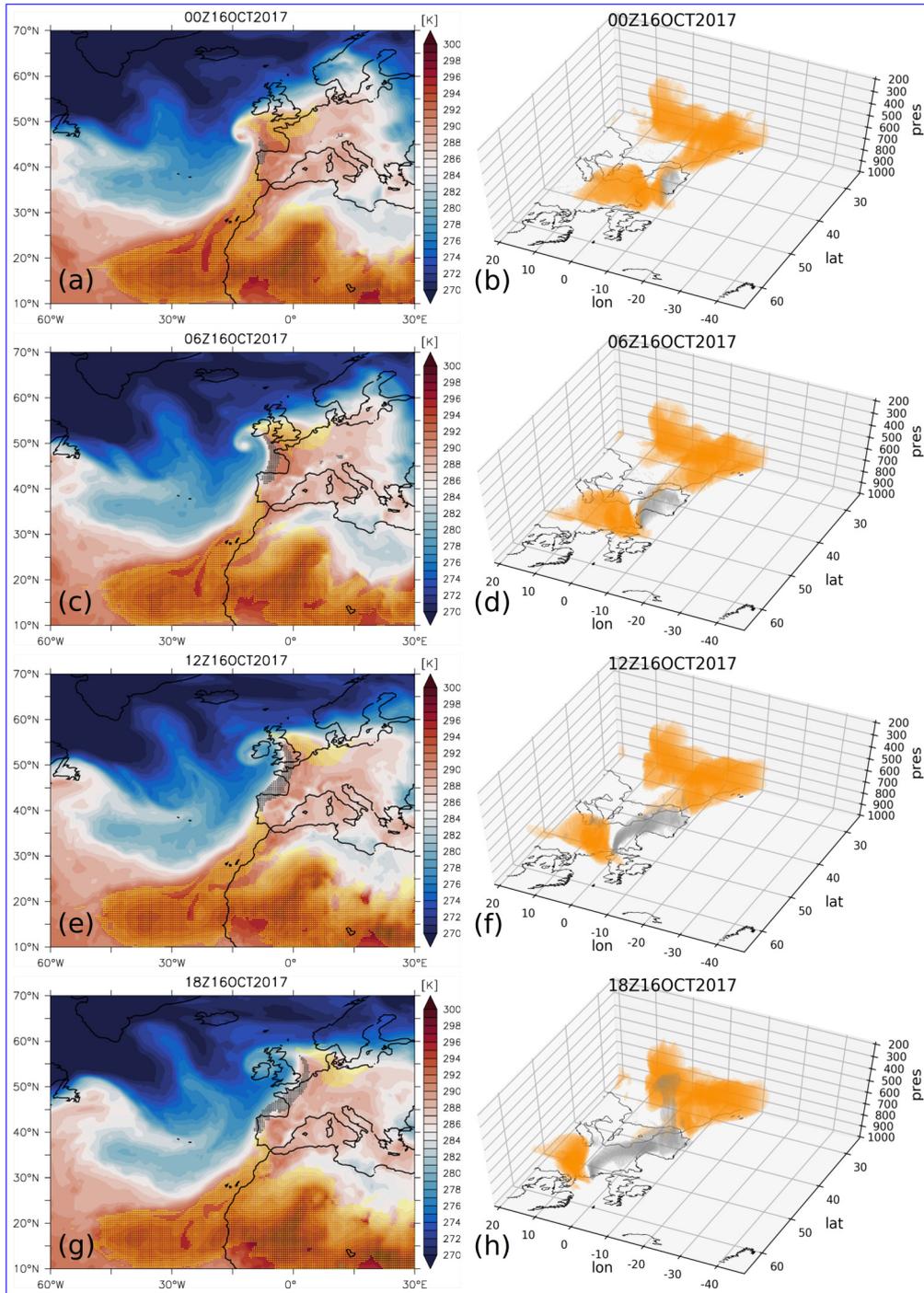


Figure 3. IFS temperature (in K; color shaded), dust with mass mixing ratio exceeding $80 \mu\text{g}/\text{kg}$ (orange shaded) and CO with volume mixing ratio exceeding 200 ppbv (grey shaded) at 850 hPa during the period from 00:00Z on 16 October 2017 to 18:00Z on 16 October 2017 (12 h interval) (Figures 3a, 3c, 3e, 3g). Three-dimensional (longitude, latitude, pressure (hPa)) spatial distribution of IFS dust with mass mixing ratio exceeding $80 \mu\text{g}/\text{kg}$ (orange shaded) and IFS CO with volume mixing ratio exceeding 200 ppbv (grey shaded) during the same period (Figures 3b, 3d, 3f, 3h).

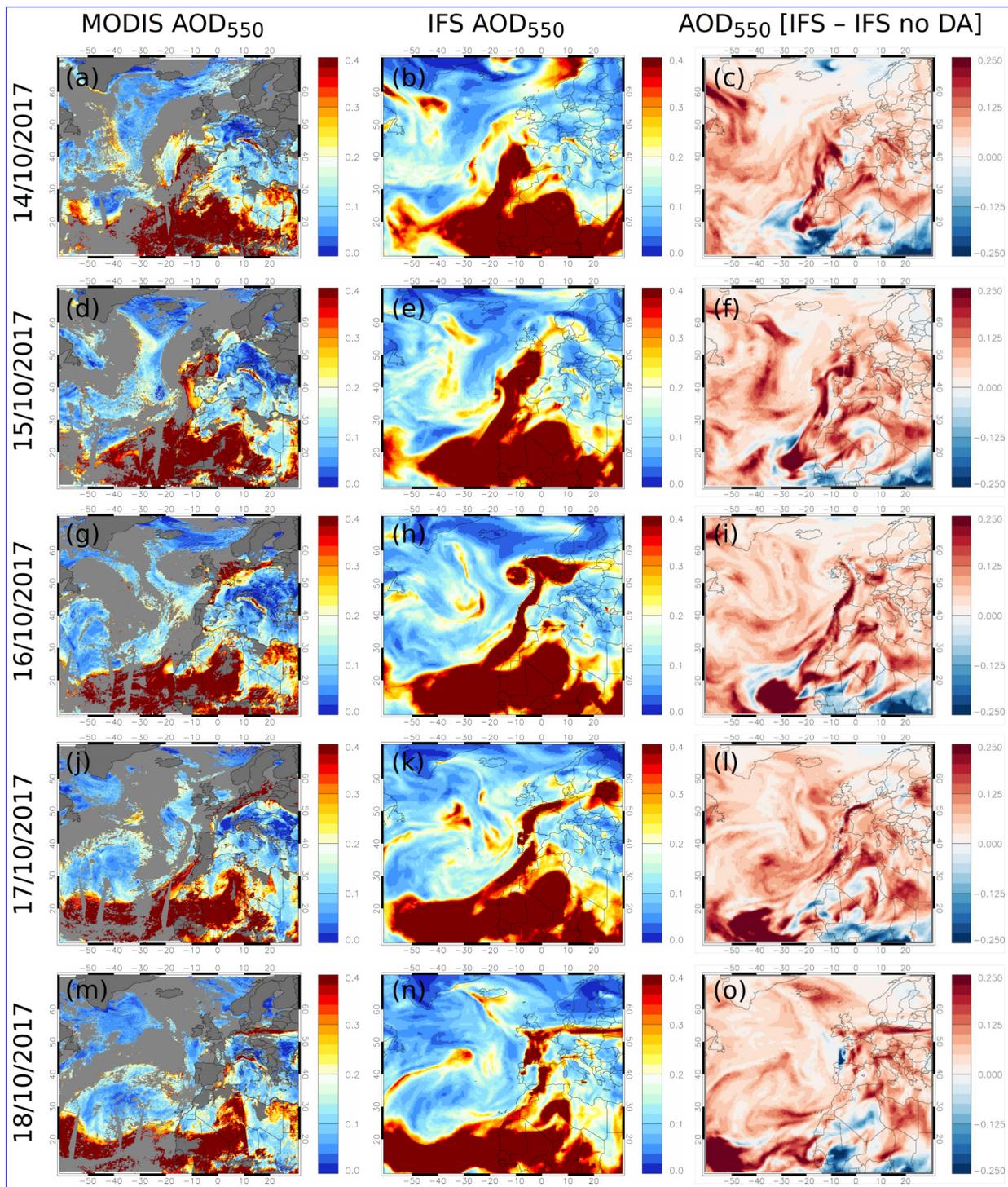


Figure 4. MODIS/Terra and Aqua Collection 6.1 AOD₅₅₀ during the period from 14 October 2017 to 18 October 2017 (Figures 4a, 4d, 4g, 4j, 4m). IFS AOD₅₅₀ (Figures 4b, 4e, 4h, 4k, 4n) and differences between IFS and IFS no DA AOD₅₅₀ (Figures 4c, 4f, 4i, 4l, 4o) at 12:00Z of the respective day.

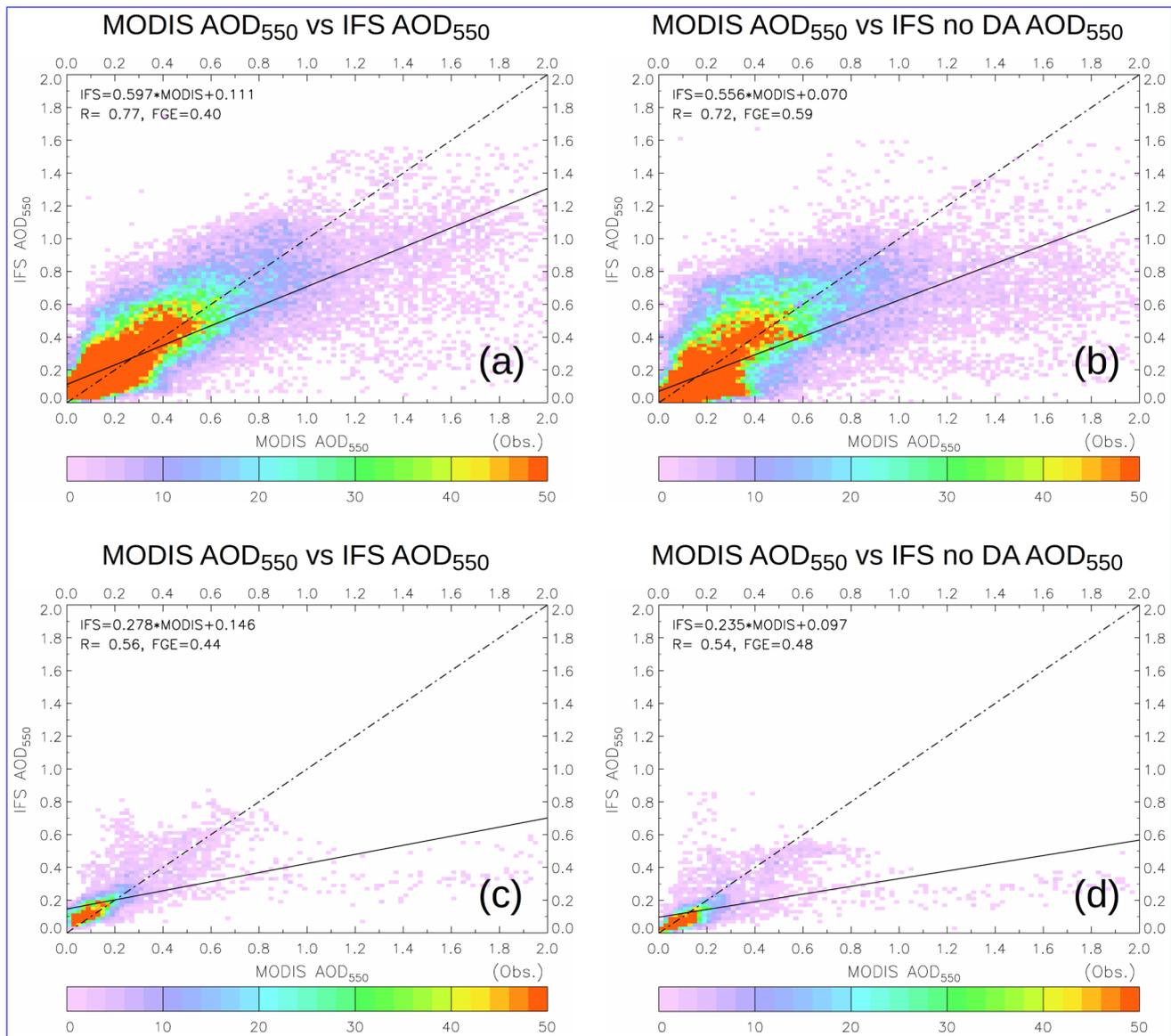


Figure 5. Comparison of spatially and temporally collocated IFS and MODIS/Terra and Aqua Collection 6.1 AOD₅₅₀ data for: the whole domain of Fig. 4, and IFS including data assimilation (a), IFS without data assimilation (b); and the box region appearing in Fig. 9d, and IFS including data assimilation (c), IFS without data assimilation (d). The examined dates are those appearing in Fig. 4. The color scale corresponds to the number of IFS-MODIS collocation points that fall within 0.02 x 0.02 AOD₅₅₀ bins. The solid line is the regression line of the IFS-MODIS data and the dashed-dotted line is the 1:1 line. The slope and the intercept of the regression line, the correlation coefficient (R) and the fractional gross error (fge / FGE) are also shown.

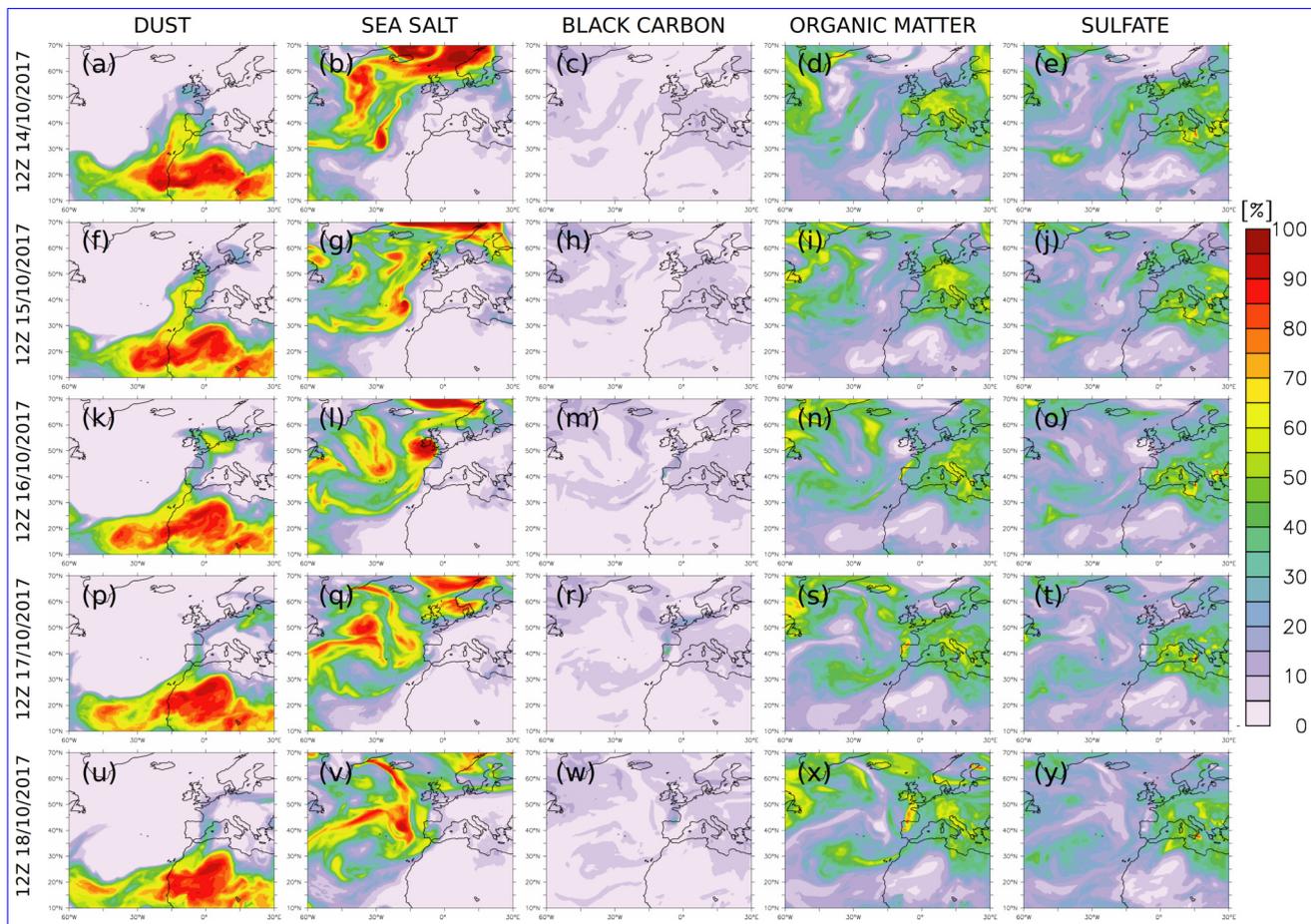


Figure 6. Percentage contribution of dust (Figures 6a, 6f, 6k, 6p, 6u), sea salt (Figures 6b, 6g, 6l, 6q, 6v), black carbon (Figures 6c, 6h, 6m, 6r, 6w), organic matter (Figures 6d, 6i, 6n, 6s, 6x) and sulfate (Figures 6e, 6j, 6o, 6t, 6y) aerosols to the IFS total AOD₅₅₀ values appearing in Fig. 4.

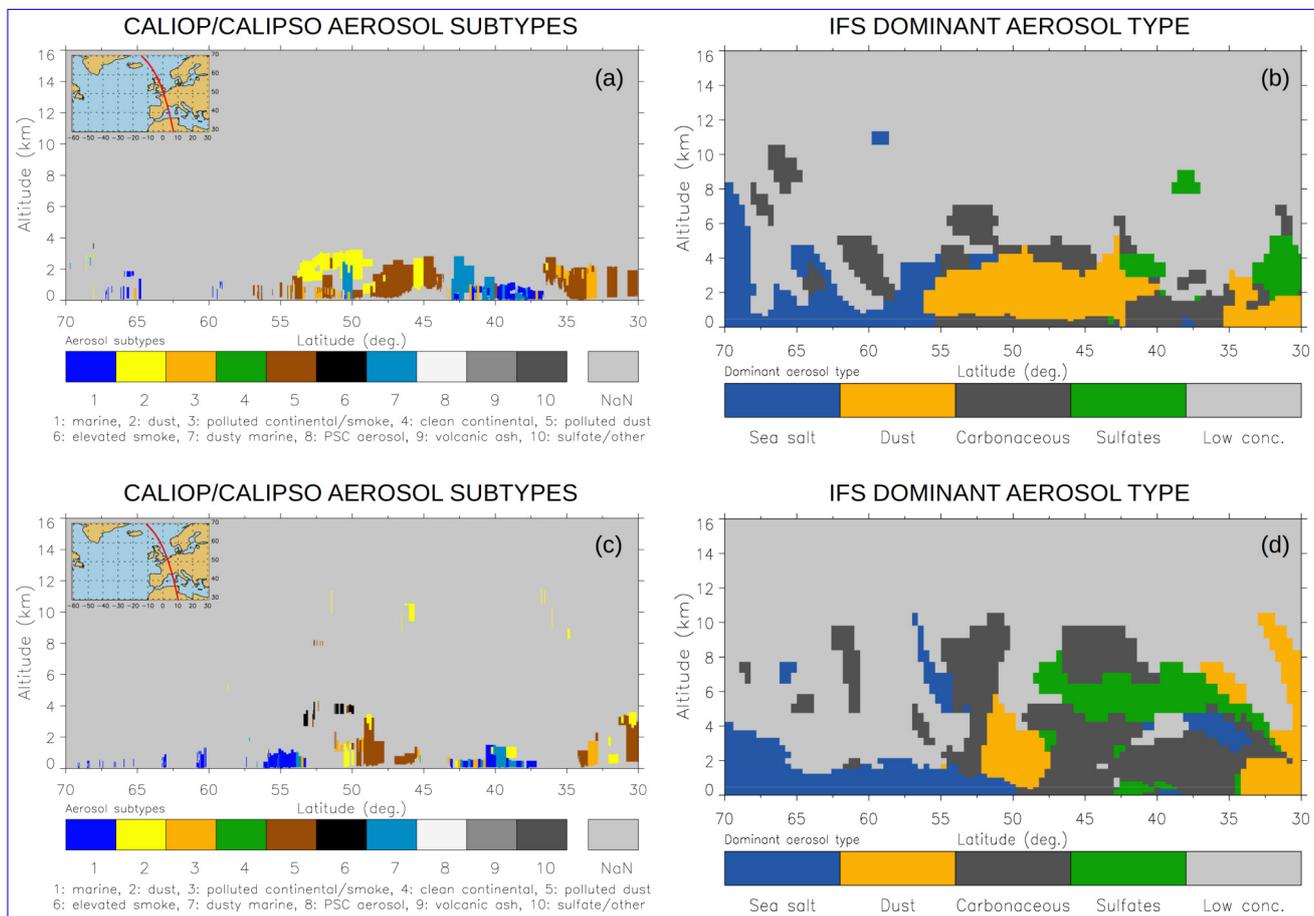


Figure 7. Vertical cross-sections of CALIOP/CALIPSO aerosol subtypes and IFS dominant aerosol type on 12Z 15 October 2017 (a, b) and on 12Z 17 October 2017 (c, d). The embedded maps in Figs. 7a and c depict the respective CALIOP/CALIPSO tracks.

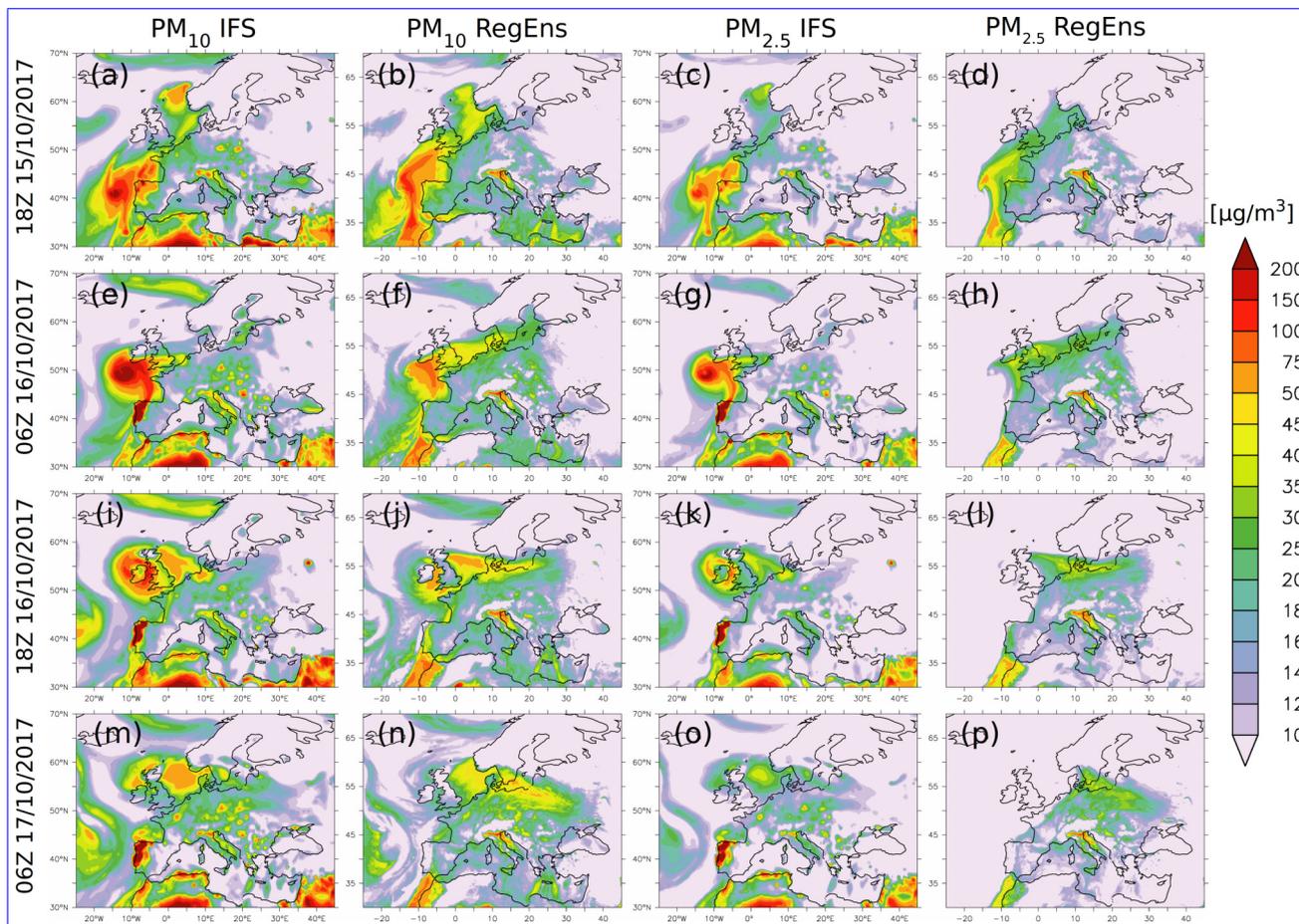


Figure 8. PM₁₀ surface concentrations (µg/m³) for IFS (Figures 8a, 8e, 8i, 8m) and RegEnS (Figures 8b, 8f, 8j, 8n) for the period from 18:00Z on 15 October 2017 to 06:00Z on 17 October 2017. PM_{2.5} surface concentrations (µg/m³) for IFS (Figures 8c, 8g, 8k, 8o) and RegEnS (Figures 8d, 8h, 8l, 8p) for the same period.

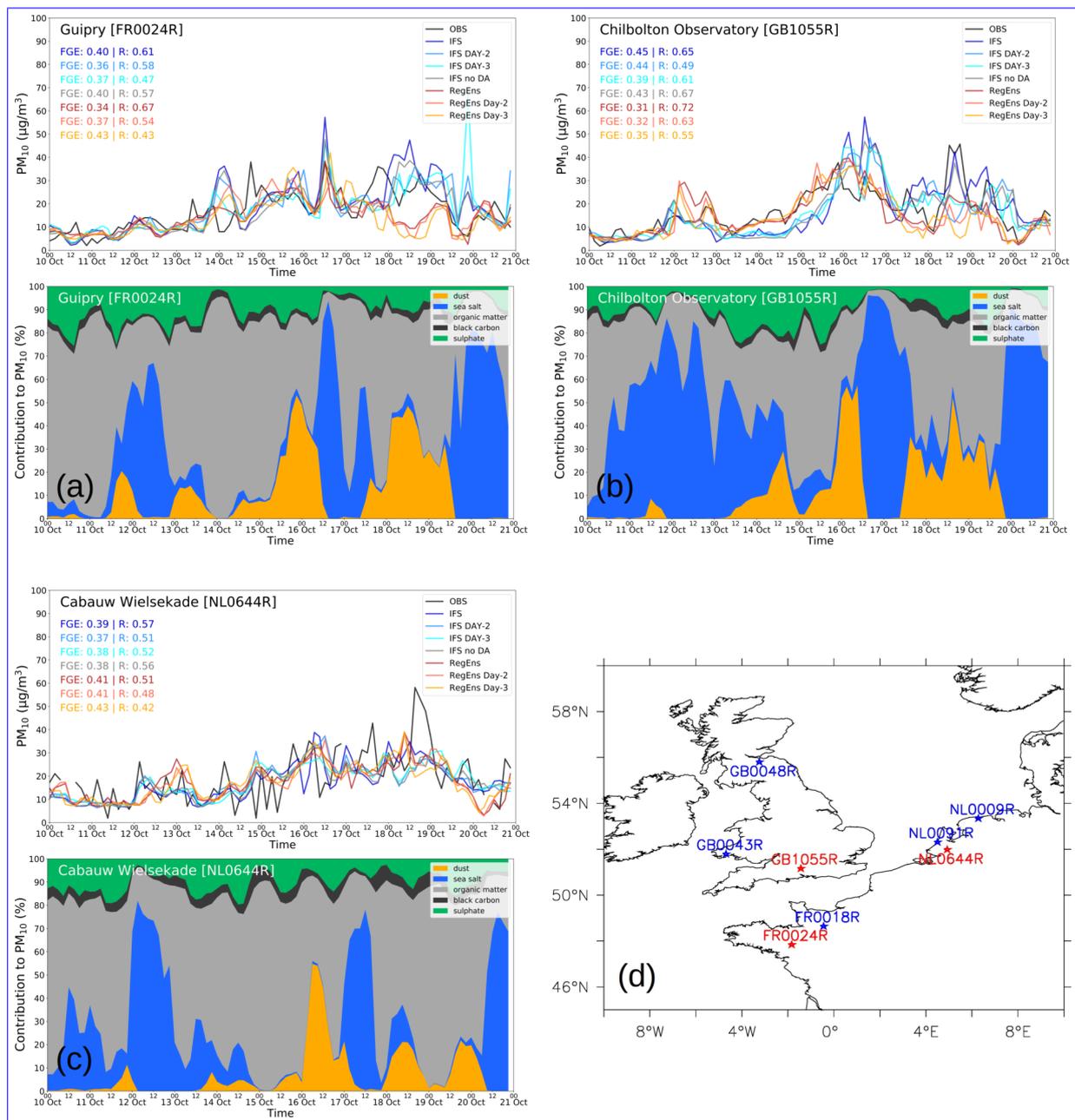


Figure 9. Observed (black), IFS (blue), IFS day-2 (light blue), IFS day-3 (aqua), IFS no DA (grey) and RegEnS (red), RegEnS day-2 (light red) and RegEnS day-3 (orange) 3-hourly timeseries of PM_{10} surface concentrations ($\mu\text{g}/\text{m}^3$) (top), and percentage (%) contribution of aerosol type to IFS PM_{10} surface concentrations (bottom) for Guipry (FR0024R), France (a); Chilbolton Observatory (GB1055R), Great Britain (b); Cabauw Wielsekade (NL0644R), the Netherlands (c). The locations of the presented EMEP stations in the manuscript (red) and the Supplement (blue) are also shown (d).

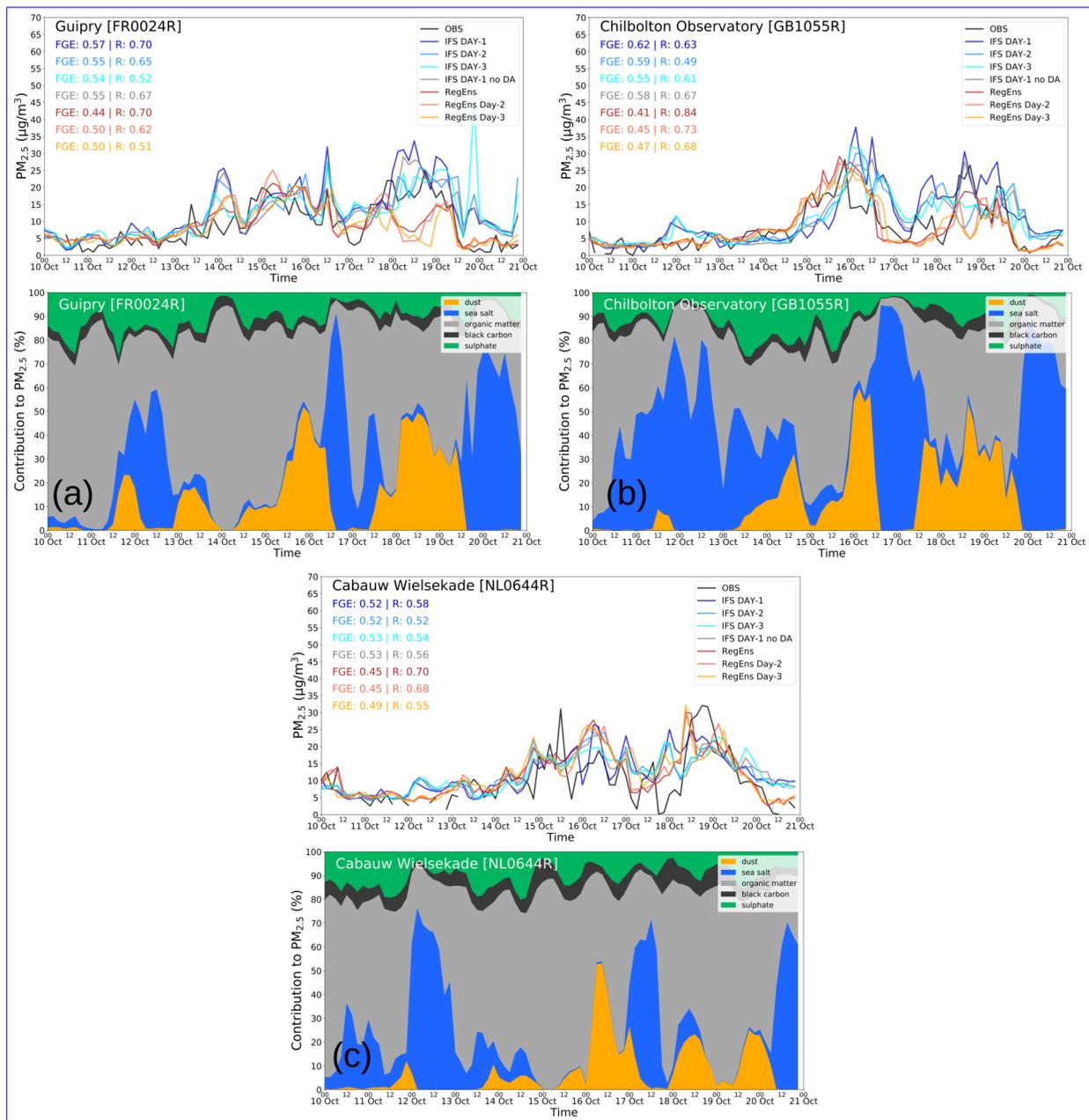


Figure 10. Observed (black), IFS (blue), IFS day-2 (light blue), IFS day-3 (aqua), IFS no DA (grey) and RegEns (red), RegEns day-2 (light red) and RegEns day-3 (orange) 3-hourly timeseries of $PM_{2.5}$ surface concentrations ($\mu g/m^3$) (top), and percentage (%) contribution of aerosol type to IFS $PM_{2.5}$ surface concentrations (bottom) for Guipry (FR0024R), France (a); Chilbolton Observatory (GB1055R), Great Britain (b); Cabauw Wielsekade (NL0644R), the Netherlands (c).

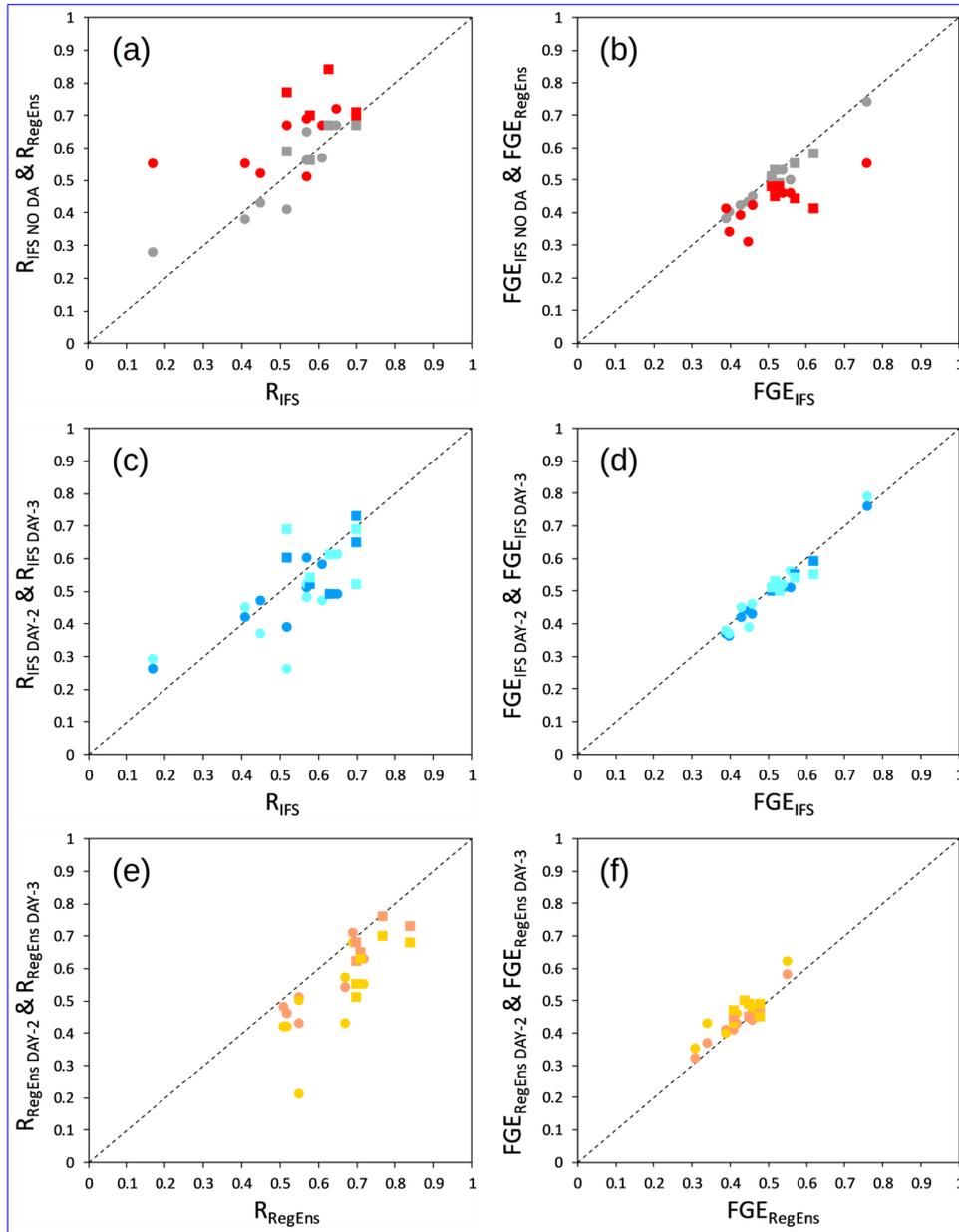


Figure 11. (a) Comparison between the correlation coefficients IFS vs OBS (\overline{R}_{IFS}) and IFS no DA vs OBS ($\overline{R}_{IFS\ NO\ DA}$) (grey symbols), as well as between IFS vs OBS (\overline{R}_{IFS}) and RegEns vs OBS (\overline{R}_{RegEns}) (red symbols) for PM_{10} (circles) and $PM_{2.5}$ (squares). (b) Same as (a) but for FGE. (c) Comparison between the fge-of-correlation coefficients IFS vs OBS (\overline{fgeR}_{IFS}) and IFS no-DA-DAY-2 vs OBS ($\overline{fgeR}_{IFS\ NO\ DA\ DAY-2}$) (grey-light blue symbols), as well as between the fge-of-IFS vs OBS (\overline{fgeR}_{IFS}) and IFS DAY-3 vs OBS ($\overline{R}_{IFS\ DAY-3}$) (aqua symbols) for PM_{10} (circles) and $PM_{2.5}$ (squares). (d) Same as (c) but for FGE. (e) Comparison between the correlation coefficients RegEns vs OBS (\overline{R}_{RegEns}) and RegEns DAY-2 vs OBS ($\overline{R}_{RegEns\ DAY-2}$) (light red symbols), as well as between RegEns vs OBS (\overline{R}_{RegEns}) and RegEns DAY-3 vs OBS ($\overline{R}_{RegEns\ DAY-3}$) (orange symbols) for PM_{10} (circles) and $PM_{2.5}$ (squares). (f) Same as (e) but for FGE. The results for PM_{10} ($PM_{2.5}$) are obtained from the 8 (5) EMEP stations described in Table 1.