Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-42
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 27 February 2018

Atmospheric Chemistry and Physics
Discussions
Open Access

Status and future of Numerical Atmospheric Aerosol Prediction with a focus on data requirements

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Abstract. Numerical prediction of aerosol particle properties has become an important activity at many research and operational weather centres due to growing interest from a diverse set of stakeholders, such as air quality regulatory bodies, aviation and military authorities, solar energy plant managers, providers of climate services, and health professionals. The prediction of aerosol particle properties in Numerical Weather Prediction (NWP) models faces a number of challenges owing to the complexity of atmospheric aerosol processes and their sensitivity to the underlying meteorological conditions. Errors in aerosol prediction concern all processes involved in the aerosol life cycle. These include errors on the source terms (for both anthropogenic and natural emissions), errors directly dependent on the meteorology (e.g., mixing, transport, scavenging by precipitation), as well as errors related to aerosol chemistry (e.g., nucleation, gas-aerosol partitioning, chemical transformation and growth, hygroscopicity). The main goal of current research on aerosol forecast consists in prioritizing these errors and trying to reduce the most important ones through model development and data assimilation. Aerosol particle observations from satellite and ground-based platforms have been crucial to guide model development of the recent years, and have been made more readily available for model evaluation and assimilation. However, for the sustainability of the aerosol particle prediction activities around the globe, it is crucial that quality aerosol observations continue to be made available from different platforms (space, near-surface, and aircraft) and freely shared. This white paper reviews current requirements for aerosol observations in the context of the operational activities carried out at various global and regional centres. Some of the requirements are equally applicable to aerosol-climate research. However, the focus here is on the global operational prediction of aerosol properties such as mass concentrations and optical parameters. Most operational models are based on bulk schemes that do not predict the size distribution of the aerosol particles. Others are based on a mix of "bin" and bulk schemes with limited capability to simulate the size information. However the next generation of aerosol operational models will have the capability to predict both mass and number density which will provide a more complete description of the aerosols properties. A brief overview of the state-of-the-art is provided with an introduction on the importance of aerosol prediction activities. The criteria on which the requirements for aerosol observations are based are also outlined. Assimilation and evaluation aspects are discussed from the perspective of the user requirements.

1 Introduction

Over the last two decades, the concept of global observing systems and the importance of defining user requirements for the purpose of monitoring and forecasting elements of the Earth System has gained momentum. This also applies to atmospheric composition in general and aerosol in particular with the studies of Barrie et al. (2004) for atmospheric composition monitoring, Reid et al. (2011) for operational aerosol forecasting, Benedetti et al. (2011) for operational verification of aerosol properties, and Colarco et al. (2014) on the use of Earth Observing System data for aerosol operational systems. Indeed, at the time of writing this document, there are at least nine operational centres producing and distributing real-time global aerosol forecasting products, including: ECMWF Copernicus Atmosphere Monitoring Service (CAMS), Japan Meteorological Agency (JMA), NASA Global Modelling and Assimilation Office (GMAO), NOAA National Centre for Environmental Prediction (NCEP), US Navy's Fleet Numerical Meteorology and Oceanography Centre (NREL/FNMOC), UK Met Office, Météo-France and the Finnish Meteorological Institute (FMI). Barcelona Supercomputing Center (BSC) is not an operational centre but provide operational aerosol forecasting products as well. Each of these centres has its own internal requirements for data to support data assimilation, verification and development of their aerosol forecasting programs. Commissioned by the World Meteorological Organization (WMO), this document outlines the requirements of the aerosol prediction system developers (the data "users" in this context). It has been compiled through consultation with experts in aerosol modelling, assimilation and evaluation both from the operational centres and the aerosol research community. Clearly establishing these requirements is important for many issues also involving the provision of aerosol variables and the evolution towards future technical requirements.

1.1 Context and needs of the numerical atmospheric composition prediction community

Numerical atmospheric aerosol prediction (NAAP) is still an activity in its infancy. It can be seen as a sub-component of the larger and far more mature field of numerical weather prediction (NWP) As such, it is reasonable to expect that NAAP will follow best practices set up by the NWP community. This includes in particular best practices in using and setting requirements for observational data. Just as there are requirements for radiosonde releases and weather station data transmission, one would expect similar considerations for parameters such as $\mathrm{PM}_1 0$ (total mass of particles with diameter less than 10 μm), $\mathrm{PM}_2.5$ (total mass of particles with diameter less than 2.5 μm) and other key parameters such as Aerosol Optical Depth (AOD), extinction coefficient, mass concentrations of individual chemical components, and light scattering and absorption coefficients. To a large degree this type of data is already being collected in many countries around the world and inter-calibration procedures are in place in existing surface networks. There are, however, a number of unique challenges facing the NAAP community that should be addressed and integrated in the development of relevant global aerosol data streams. There is a long history of reporting and sharing meteorological data because it is understood to be of mutual benefit to all parties in the exchange and, weather being considered an "act of nature" there is less political motive behind data policies. Atmospheric composition data, however, is often related to air quality through anthropogenic emissions of pollutants and thus has local regulatory or even international treaty ramifications. There can subsequently be some local hesitance to report unfavourable data, or at the least to provide additional funding to ease its distribution. One exception is dust storms, and indeed reporting of dust observation and prediction is more mature than any other aerosol species, even though there are only a few ground stations in key source areas. Compositional data collection also requires expensive and, often, difficult to calibrate equipment. While NWP has suffered at times with diversity in, for example, commercial radiosonde providers and instrument efficacy, aerosol measurement has considerably more degrees of freedom in its measurement technology, overall maintenance, and reporting. While institutions such as the World Meteorological Organization (WMO), the United States Environmental Protection Agency (EPA) or the European Environment Agency (EEA) set benchmark levels for air quality monitoring, they are by no means universally applied. The research community is nevertheless making a huge effort to intercompare and standardize their measurements. The ability to report with a given timeliness is related to measurement technology. A host of potential variables can be generated relating to mass, composition, optical properties, or microphysics. Deployed instruments and their locations are also constantly evolving. The authors of this paper are keenly aware of the difficulties associated with aerosol measurements and the efforts made to improve these. The "requirements" or recommendations made herein should not be interpreted as criticisms of the existing observing system but as a mean to move forward. They are not meant to introduce more rigidity and should be interpreted practically. Given the early state of the field and diversity in development approaches and customer requirements at aerosol prediction centres, the community requires flexibility as it finds its way. Regardless of data type, whether in situ or from remote sensing, there are three guiding principles that should be considered.

1. Data should be easily accessible, distributable, and for baseline quantities, encoded into a similar format. Currently data distribution is diffuse and potential users have difficulty maintaining and evaluating global scale data outside of the largest and most consistent networks (for example the NASA Aerosol Robotic Network-AERONET sun photometer dataset). While long term sites are preferred, the operational reality has been for a reduction in support for key supersites, such as Atmospheric Radiation Measurement (ARM) or Global Atmospheric Watch (GAW). Thus, future data distribution models could mimic meteorological data, where observations are broadcast and consolidated for use (e.g., 6 or 12 hourly $PM_2.5$ or PM_10 data). However, care must be taken to avoid ongoing legacy issues in the current broadcast system.

2. Timeliness requirements also vary by center. Based on the consensus of centers, 3 hour latency is preferred, and 6 hours is adequate, especially for satellite products. There is nevertheless value in 12 hour or even multi day delivery for verification purposes, including surface particulate matter monitoring. Timeliness should be a goal, but not necessarily a requirement. This is especially true for compositional data requiring laboratory work for analysis.

3. Realistic error bars or error models must be provided. The operational community can easily cope with uncertain data, provided that uncertainty is known. Indeed, error tolerances are strongly customer related.

Mindful of these considerations, specific issues and definitions of user requirements are addressed in the following subsections. Note that in this paper no mention is made of volcanic ash aerosols. While the prediction of this type of aerosol is essential for numerous applications, we believe that there is a need for a separate study dealing with specific requirements for the prediction of volcanic ash aerosols. Several communities are dealing with this topic, for example the Global Atmospheric Watch (GAW) Scientific Advisory Group (SAG) on Volcanic Ash, the GAW SAG on Applications, the global aerosol lidar network GALION, and others. The AEROSOL Bulletin 3 available from WMO provides an overview of current efforts on this topic.

1.2 The nature of user requirements

The notion of user requirements implies that the specific technology or science application has an underlying group or community that has an interest in using the data, be it data from an observational platform or from a model. Communities use the data for their applications, and this (implicitly or explicitly) sets the requirements. One of the principles behind user requirements implies that data requirements should be put forward by the relevant communities independently of the current technologies and systems available, with the overarching goals of supporting the applications of the community in question, for example weather prediction, ocean modeling, climate investigation etc. Specifically for observation requirements, no consideration is given to what type of instruments, observing platforms or data processing systems are necessary or even possible to meet them. Even though in practice, it is not possible to make user requirements completely technology-free and current availability of technology influences their formulation, it is a useful exercise to understand data gaps and also to establish if new observing systems can meet all or part of the user requirements. This process of formulating user requirements establishes also a direct link between model developers and data providers, which is extremely important. Many data products that are provided by environmental agencies or individual scientists, end up not being in the model development/ assimilation/ assessment loop as they do not correspond with what is needed by the modellers (e.g., in terms of accessibility, timeliness, quality, or uncertainty). Vice versa, often model developers have unrealistic expectations, do not specify their priorities and end up using only a sub-set of available observations. Dialogue between these two communities is what ultimately fosters progress on both sides. The requirements for observations are usually given in terms of the following criteria: (i) resolution (horizontal and vertical and sometimes temporal), (ii) sampling (horizontal and vertical), (iii) frequency (how often a measurement is taken in time), (iv) timeliness (i.e., availability), (v) repetition cycle (how often the same area of the globe is observed), and most importantly (vi) uncertainty either related to the actual instrument accuracy and/or to the algorithm used to perform the retrieval in case of derived observations (for example aerosol optical depth or total column ozone). Additionally, the user must specify what physical or chemical variables should be measured.

Resolution and sampling differ in that resolution relates to the area and time period a measurement is representative of, whereas sampling indicates the distance between two successive measurements both in space and time. Frequency is related to the temporal sampling of an instrument whereas repetition gives a measure of how often the same location is observed. For example, an instrument on a polar orbiting satellite may have very high frequency but low repetition.

Uncertainty can be divided into accuracy, which relates to the bias of the measurement, and precision, which relates to the random error. For example in the presence of biased observations, averaging more observations does not generally improve the accuracy, but may improve the precision. For each application, it is generally accepted that improved observations in terms of resolution, sampling, frequency and accuracy, etc. against some baseline are generally more useful than coarser, less frequent and less accurate counterpart observations. The latter, however, could still be useful. Some of the criteria may come into play depending on the particular area of application. For example, timeliness is a criterion which is not included in the requirements for climate research whereas due to the constraints on the timely delivery of the forecasts, it is a crucial parameter for operational prediction and assimilation. The usefulness of an observation is dependent on the specific application and its availability. This is specified in the requirements by adding three values per criteria: the “goal”, the “threshold” and the “breakthrough”. The goal is the value above which further improvement of the observation would not bring any significant improvement to the application. Goals may evolve depending on the progress of the application and the capacity to make better use of the observations. The threshold is the value below which the observation has no value for the given application. An example of threshold requirement for assimilation is, for example, the timeliness of the data: observations that are delivered beyond a certain time (normally three to six hours for near-real time NWP applications) cannot be used in the analysis. The breakthrough is a value in between the goal and the threshold that, if achieved, would result in a significant improvement for the application under consideration. Of these three parameters the most elusive is the breakthrough because its value may change more drastically than the other two with system developments. While the usefulness class of requirement is conceptually straightforward it is less so functionally and consequently can have an arbitrary nature in a rapidly developing field such as NAAP. Thus, while this document will provide examples of usefulness, there is a hesitation to be overly specific at this time. In particular breakthrough and goal values for different variables are not independent: accurate measurements of one variable may lower the usefulness of another less accurately-measured variable because the variables are related in the model. For instance AOD measurements become less valuable if measurements of the extinction coefficient become available with the required sampling and accuracy.

1.3 Rolling Review of Requirements and Task Team on Observational Requirements and Satellite Measurements

The WMO has developed a framework for different thematic areas such as Global Numerical Weather Prediction, High-resolution Numerical Weather Prediction, Nowcasting and Very Short Range Forecasting, Ocean applications, and Atmospheric Chemistry, among others, to be reviewed periodically in terms of design and the implementation of various observing systems, using as guidance the user requirements set-out by the relevant community (Barrie et al., 2004). This process is called the rolling review of requirements (RRR) and it involves several steps. For each application area, these steps are as follows: (i) a review of "technology-free" user requirements (i.e., not taking into account the available technology) for observations in one of the thematic areas; (ii) a review of current and future observing capabilities (space-based and surface-based); (iii) a critical review of whether the capabilities meet the requirements; and finally (iv) a statement of guidance based on the outcomes of the critical review. This statement of guidance is often called gap analysis as it shows whether the current observing system is suitable for the given application and what is needed in the future observing system in order for it to meet the requirements set out by the user community. To facilitate this process, the WMO maintains an online database on user requirements and observing system capabilities called Observing Systems Capability Analysis and Review tool (OSCAR). Details on the RRR are provided in Eyre et al. (2013), and references therein.

Recently, the WMO GAW set up an ad-hoc Task Team on Observational Requirements and Satellite Measurements as regards Atmospheric Composition and Related Physical Parameters (TT-ObsReq, http://www.wmo.int/pages/prog/arep/gaw/TaskTeamObsReq.html) to review the user requirements specifically for atmospheric composition. Application areas related to atmospheric composition include: (i) Forecasting Atmospheric Composition which covers applications from global to regional scales ($\approx$10 km and coarser) with stringent timeliness requirements (NRT) to support operations such as sand and dust storm and chemical weather forecasts, (ii) Monitoring Atmospheric Composition which covers applications related to evaluating and analysing changes (temporally and spatially) in atmospheric composition regionally and globally to support treaty monitoring, climatologies and re-analyses, assessing trends in composition and emissions/fluxes, and to better understand processes, using data of controlled quality (and with less stringent time requirements than needed for NRT). (iii) Providing Atmospheric Composition information to support services in urban and populated areas, which covers applications that target limited areas (with horizontal resolution of a few km or smaller) and stringent timeliness requirements to support services related to weather/climate/pollution, such as air quality forecasting.

The WMO GAW TT-ObsReq analysed the role of atmospheric composition observations also in support of the other WMO application areas (http://www.wmo.int/pages/prog/www/OSY/GOS-RRR.html). After the Second Workshop of the TT-ObsReq (12-13 August 2014, Zurich), the committee identified key parameters needed for Forecasting Atmospheric Composition. For aerosols these parameters were: aerosol mass, size distribution (or at least mass in three fraction sizes: up to 1, 2.5 and 10 micron as it is common practice in air quality, speciation and chemical composition, AOD at multiple wavelengths, absorption AOD (AAOD), ratio of vertically integrated mass to AOD, vertical distribution of aerosol extinction). Some of the parameters outlined for Monitoring Atmospheric Composition may also be relevant to the operational prediction of aerosol particle properties, which is one of the application areas (Forecasting Atmospheric Composition) and is the focus of this study. Because recommendations from the committee are technology-free, they differ slightly from those identified by the Scientific Advisory Group on Aerosol (GAW report 227), which limits recommendation to variables that can be directly measured.

Requirements are outlined based on what is needed for the fundamental components of an aerosol prediction system which are: (i) modelling processes (aerosol particles emission and removal), (ii) data assimilation (when present), and (iii) model evaluation. Section 2 briefly presents current operational and pre-operational aerosol systems both at the global and the regional scales. Section 3 describes the data needs and the requirements for emission and removal processes, section 4 outlines those for the assimilation component, and finally section 5 describes those related to model evaluation. Section 6 summarizes those data needs and includes some final thoughts.

2 Aerosol Prediction Models

Several centres with operational capabilities are currently running aerosol prediction systems. These are BSC, ECMWF, FNMOC/NRL, JMA, Met Office, NASA, NCEP, on the global level and BSC, the Chinese Meteorological Agency (CMA), the Korean Meteorological Agency (KMA), the Institut national de l'environnement industriel et des risques (INERIS), Météo-France, the Deutscher Wetterdienst (DWD), to mention a few on a regional level. Some centres such as BSC, the UK Met Office, NCEP, Météo-France and FMI currently run operational dust forecasting systems, and have also developed capabilities for other aerosol species (sea salt, carbonaceous aerosols, sulphates). Most of these systems also have assimilation capabilities. A detailed description of the individual models is beyond the scope of this paper. For a review of the current systems that provide aerosol forecasts, some with focus on dust, see for example Benedetti et al. (2014) and Sessions et al. (2015).Ensemble systems are presented in Rubin et al. (2016) and Di Tomaso et al. (2017). An overview of regional aerosol forecasting systems can be found in Menut and Bessagnet (2010); Kukkonen et al. (2011); Zhang et al. (2012a, b); Baklanov et al. (2014).

In the rest of the paper, we will mainly focus on requirements for global models, acknowledging that regional (i.e., limited-area) models may have different sets of requirements, including some for boundary conditions. Regional ground-based networks can for example address some of those needs while not providing sufficient coverage for global models. Global observations can be of use also for regional applications but the requirement on the resolution, for example, may differ from that of a global model. In general most of the requirements below will apply to both global and regional models.

2.1 Multi-model consensus ensemble predictions

In recent years, aerosol forecasting centres have been turning to ensemble prediction to describe the future state of the aerosol fields from a probabilistic point of view. Multi-model product consensus ensembles of deterministic aerosol forecast models have been developed to alleviate the shortcomings of individual models while offering an insight on the uncertainties associated with a single-model forecast. Examples include the International Cooperative for Aerosol Prediction (ICAP) Multi Model Ensemble (ICAP-MME, Sessions et al. (2015) (http://www.nrlmry.navy.mil/aerosol/) for global aerosol forecasts and the WMO Sand and Dust Storm Warning Advisory and Assessment System (SDS-WAS) North African and Middle East Regional Node for regional dust forecasting (http://sds-was.aemet.es/; Terradellas et al. (2016)). The ICAP initiative itself has demonstrated that simply collecting different forecasts in a single database and generating web pages with common plotting conventions is an effective tool for developers to assess and improve their forecasting systems. Use of ensemble forecast techniques is especially relevant for situations associated with unstable weather patterns, or in extreme conditions. Ensemble approaches are also known to have more skills at longer ranges (> 3 days) where the probabilistic approach provides more reliable information than a single model run due to the model error increasing over time. Moreover an exhaustive comparison of different models with each other and against multi-model products as well as observations can reveal weaknesses of individual models and provide an assessment of model uncertainties in simulating the aerosol cycle. Multi-model ensembles also represent a paradigm shift in which offering the best product to the users as a collective scientific community becomes more important than competing for achieving the best forecast as individual centres. This new paradigm fosters collaboration and interaction, and ultimately results in improvements in the individual models and in better final products.

3 Modeling of aerosol particle emission and removal processes

3.1 General concepts

Modeling of aerosol particle sources and sinks are of uppermost importance, because these processes largely control the spatio-temporal distributions of aerosol particle concentrations and size distributions. In addition, in polluted environments, uncertainties are dominated by emissions whereas in remote regions aerosol processes control the uncertainty. For a given source strength, sinks also control the atmospheric residence times of aerosol particles, which is in turn a key indicator of long-range transport of aerosol species. A good representation of aerosol particle sources and sinks is particularly important to determine the overall analysis and forecast of particle mass, surface area, and number concentrations in regions with few observations for data assimilation. A discrepancy in aerosol sources and/or sink processes can cause a systematic drift in aerosol particle concentrations and AOD over the forecast range in a forecasting system with data assimilation. This is because often the data assimilation corrects for the bias in sources and/or sinks. This correction is often not retained in the subsequent forecast integration due to the fact that the model does not represent the emission/removal processes adequately. For this reason, it is useful to set user requirements also for source and sink observations of aerosol particles. Efforts to formulate aerosol DA with emission fluxes as well as / instead of mixing ratios as a control variable might have a role to play in correcting these forecast drifts, although such observations would remain important constraints in such a framework.

It is appropriate to differentiate sources of aerosols and aerosol precursors that are directly emitted by human activities from those (natural or anthropogenic) emissions that depend on natural processes. User requirements for directly-emitted anthropogenic emissions can be articulated around the following criteria: accuracy, spatial resolution, temporal resolution, speciation, and aerosol size distribution. User requirement for emissions that depend on meteorological processes also include requirements on key meteorological and environmental quantities that control such emissions, for example wind and surface conditions or any other parameters that may lead to aerosol formation.

3.2 User requirements for emissions: anthropogenic and biogenic aerosols

What is generally perceived as anthropogenic air pollution is in fact a result of complex and poorly understood photochemical processing as well as emissions from point and area sources. Often, anthropogenic emissions are taken to be those associated with domestic, industrial, and mobile sources. However, agricultural emissions, including fertilizers and open maintenance burning, are inconsistently included in the terms biogenic and anthropogenic, respectively. This ambiguity can be initially handled by accepting that, from an aerosol point of view, it is all a single class of processes and “anthropogenic” and “biogenic” emissions follow a similar processing rubric in models. Gridded emissions inventories are commonly generated for primary particles (e.g., sea salt, dust, primary organic matter (POM), and black carbon (BC)). Sulfates, nitrates, other inorganics, secondary organic aerosol (SOA), and black carbon (BC) are supplemented by emissions of key gases important in secondary aerosol particle production (e.g., SO_2, NOx, ammonia, isoprene, alkenes, aromatics, terpenes, etc.). These inventories are the result of large scale land classification maps, fuel inventories, and transportation corridor databases. Individual “source” classifications vary by study author, but often include power production, heavy industry/smelting, domestic and biofuel, mobile sources, road dust, agricultural field emissions, agricultural/domestic stack and burn piles as well as plant emissions of such species as isoprene and terpenes. We hold as distinct larger open biomass burning, including agriculture field burning.

3.2.1 Accuracy

Aerosol particle sources are usually prescribed from compiled emission inventories. Despite the efforts put in emission inventories by the community and continuous progress, there remain inherent difficulties in producing accurate inventories. This is because of a number of reasons such as the large variety of point and diffuse sources, uncertainties in emission factors, unknown or unaccounted for sources as well as the model emission approach that is applied (López-Aparicio et al., 2017). Among emission uncertainties there exist a hierarchy of errors. While point and area sources are less uncertain year after year thanks to satellite data, emission factors remain uncertain due to the impossibility to measure them in “realistic" conditions and their strong dependence on the environment.

Since the error on emission inventories automatically translates into a similar or even larger error in concentrations, a user requirement on emission uncertainties might be tempting. However it should be kept in mind that uncertainties and biases in emissions are difficult to estimate and reducing the error to a single number might not be possible. Aerosol source inversion techniques (e.g., Huneeus et al. (2012); Escribano et al. (2017)) have made some progress but are not yet at a stage where they can constrain emission inventories to better than the user requirement. Such studies can nevertheless point to regional problems in emission inventories.

3.2.2 Spatial resolution and sampling

One ideally requires emission inventories that have a resolution as good as the model resolution. For global modelling systems, this amounts to a spatial resolution and sampling of typically 50 km, although of course many benefits in modelling aerosol transport and deposition may be gained by running NWP at high resolution, even if sources are not known at that resolution. As computing power increases, it is relatively easy to increase model resolution. Sub-grid scale information in emission inventories can be use to post-process and downscale, at least statistically, the simulated model concentrations (Wang et al., 2014). New methods based for example on population density as a proxy are also being used (Mailler et al., 2017).

For these reasons, it is appropriate that global emission inventories always aim for spatial resolution and sampling that are higher than that of models at a given time (i.e., we recommend a minimum of 10 km resolution given the current state of play). Even higher resolutions (< 1km) are required for regional air quality models given that the typical scale for emissions is very small (e.g., the width of a road for surface traffic).

3.2.3 Temporal distribution and sampling

Temporal distribution of emission inventories can be critical as emission inventories need to sample the diurnal, weekly and seasonal cycles in emissions. Since some aerosol data products are only available at day (e.g., AOD retrieved in the visible part of the electromagnetic spectrum), it is important to deal with the diurnal cycle in emissions so as not to introduce biases in the simulated quantities. As modeling improves, it may become necessary to move from static gridded inventories to include feedbacks with societal (e.g., public holidays, agricultural practices) or meteorological (e.g., influence of cold spells on emissions from heating/biofuel systems or dry spells/wind on stack burning) conditions. Biogenic emissions from plants have also a strong dependency on temperature and water stress.

3.2.4 Speciation

Aerosol particle speciation in global aerosol models should be reflected in global emission inventories with a minimum of aerosol precursors such as SO_2, NH_3, NOx, and primary aerosol particles such as elemental or black carbon and primary organic carbon. Industrial dust and fly ash are often left out but can be important in some regions (as China), and should be included in user requirements. Requirements on speciation for volatile organic compounds (VOCs) are more difficult to set out because it is unclear what level of complexity is required in global aerosol models whose aim is to reproduce mass or number concentrations or optical thickness due to secondary organic aerosols (SOA). We argue here that speciation of VOCs is directly related to the complexity of the aerosol scheme considered and is more difficult to link to user requirements. This is further complicated by SOA production likely being a product of joint anthropogenic emissions. At the minimum bulk seasonal emissions of key classes of reactive VOCs are required (e.g., alkenes, aromatics, isoprene, terpenes).

3.2.5 Size distribution

Aerosol particle properties, such as size and composition, play an important role in determining the aerosol particle radiative efficiency and the ability to serve as cloud condensation nuclei, as well as in having health-related impacts. User requirements on aerosol particle mass or number size distributions translate into user requirements on aerosol particle size resolution at the emission points. Such user requirements can be expressed in several ways, i.e. on PM_10, $PM_2.5$ and PM_1 emission rates, or in combined requirements on aerosol particle mass and number emission rates for typical aerosol size ranges. Historically, the focus has been first on PM_10, then $PM_2.5$ and lastly on PM_1 both for health impacts and its connection to cloud formation. The concept itself of PM at a given size cut-off is directly linked to the availability of sampling inlets, but with more current and future instruments we can expect to have a complete information on the aerosol size distribution.

3.3 User requirements for emissions: open biomass burning aerosols

Biomass burning emissions represent a highly temporally and spatially variable source of aerosols to the atmosphere and reliable and timely estimates are a key input to air quality and atmospheric composition forecasts. Here we define open biomass burning emissions as emissions by fire consuming open vegetation in fields, grasslands or forests. Biofuel or stacked agricultural burning are included in the anthropogenic and biogenic emissions and not considered as open biomass burning. Within the International Global Atmospheric Chemistry (IGAC) project, there is at present about half a dozen advanced global aerosol models that include emissions from vegetation fires that could be included in a multi-model ensemble forecast. Only half of the IGAC-participating models are currently operational, the others remain in a research and development stage (for some, only the vegetation fire component is not operational).

Several real-time smoke forecasting products exist, and are related to satellite based active fire hot spot or burn area databases. The most established global aerosol forecasts are represented in the International Cooperative in Aerosol Prediction (ICAP). Four models include dedicated smoke treatment: CAMS (ECMWF and partners), MASINGAR (MRI-JMA), GEOS-5 (NASA), NAAPS (US Navy), where the first two use emissions from the Global Fire Assimilation System (GFAS, Kaiser et al. (2012)), the second from a similar FRP-based Quick Fire Emissions Database (QFED, Darmenov and da Silva (2013)), and the last from the hot spot-based Fire Locating and Modeling of Burning Emissions (FLAMBE) system (Reid et al., 2009).

Currently most models scale biomass burning emissions to reach acceptable values of biomass burning aerosol optical thickness close to observations (MODIS or AERONET). This scaling factor ranges from 1.7 for the Met Office Unified Model limited area model configuration over South America that was used for the SAMBBA campaign (Kolusu et al., 2015) to 1.8-4.5 for GEOS-5 (Colarco et al., 2011) and 3.4 for CAMS (Kaiser et al., 2012). In CAM5 (Tosca et al., 2013), regional scaling factors are used (Lynch et al., 2016). The need for this scaling factor arises either from possible underestimation of the biomass burning aerosol emissions or from model biases.

There are still large uncertainties in how to convert between the observed fire radiative power and the total amount of dry matter actually burned during the event. In addition when the total burnt dry matter is estimated, the conversion into chemical volatile elements only takes into account static maps of vegetation cover (Heil et al., 2010).Several studies have shown this is quite a crude assumption as fuel moisture and weather conditions (e.g temperature and humidity) can largely influence the amount of emissions (French et al., 2004, 2011). Vertical and/or horizontal resolution may also play a role.

A major challenge in modeling biomass burning aerosols (BBA) is therefore to better quantify the errors that lead to the use of these scaling factors and to improve the emission datasets and/or the models so as to reduce their use.

Extensive work in drawing user requirements has recently been done by the Interdisciplinary Biomass Burning Initiative (IBBI) and GAW APP-SAG. A draft Concept Note and Expert Recommendations for a Regional Vegetation Fire and Smoke Pollution Warning and Advisory System (RVFSP-WAS) was written, which form the basis of user requirements for biomass burning aerosols (WMO GAW Report No. 235, available at http://www.wmo.int/pages/prog/arep/gaw/documents).

3.3.1 Estimating Fire emissions

Reasonably fast and comprehensive estimates of smoke constituent emissions are derived from satellite observations of fires. There are three fundamentally different approaches:

- **Active Fire and Fire Radiative Power**:

  Vegetation fires exhibit both smoldering and flaming combustion, with flame radiometric temperatures of between 750 and 1200 K appearing dominant. Wien's Displacement Law indicates that the peak of thermal emission from bodies at these temperatures occurs in or close to the shortwave infrared (SWIR; 1.6–2.5 μm) and middle infrared (MIR;3–5 μm) atmospheric windows. Fires are typically very much more active by day than by night (Giglio, 2007; Roberts et al., 2009), and the presence of relatively strong solar reflective signals in the SWIR region by day means that active fire detection algorithms are generally focused on exploitation of the MIR signal. Many publications have outlined the basis by which elevated MIR signals are used to detect actively burning fires (Robinson, 1991). In the MIR wavelength range, the spectral radiance emitted from an open vegetation fire can be up to four orders of magnitude higher than that from the ambient temperature background, therefore allowing even extreme sub-pixel fires (e.g. covering down to 0.1 -0.01% of a pixel) to significantly affect the measured MIR pixel signal. Detection of these elevated MIR channel signals, either on the basis of spectral radiance or brightness temperature measures, is the basis of most active fire detection algorithms. However, by day solar-heating of bare ground and specular sun glints from unmasked clouds or water bodies can also increase MIR pixel signals far above those of the surroundings, and thus especially by day a series of additional optical and thermal channel spectral and/or spatial tests need to be deployed to discriminate “true" fire pixels from “false" alarms. In particular, areas that are homogeneously warm due to solar heating tend to have spatially consistent and rather similar MIR and TIR brightness temperatures whereas the presence of a sub-pixel sized fire within the FOV elevates the brightness temperature difference between these two channels (MIR-TIR)), usually over just a few pixels corresponding to the location of the active flaming and/or smouldering zone. Furthermore, whilst elevated (MIR-TIR) values also occur due to sun glint effects, such pixels also show a markedly increased VIS/NIR channel, which is not the case for fires. Based on these principles, fire detection uses a series of multispectral MIR, TIR and VIS/NIR channel tests to first discriminate “potential" active fire pixels, and then confirm which of these are “true" fire pixels.

  All these channels can be observed in real time from Geostationary sensors such as GOES (e.g., Prins et al. (1998)) and polar orbiting sensors such as AVHRR (Flasse and Ceccato, 1996) and MODIS (e.g., Giglio et al. (2013)). If a quantitative fire black body temperature is extracted, then it can be interpreted as “fire radiative power” (FRP). Under certain conditions FRP can be proportional to the biomass combustion rate (Wooster et al., 2005). Subsequently, instantaneous emission rates of various smoke constituents can be calculated with assumed emission factors from the scientific literature (Heil et al., 2010). The advantages of this approach are the immediate availability of the observations and emission estimates, even while the fires are still burning, and the relatively weak dependence on the fire type for above-ground burning. However given the uncertainties in the conversion parameters, the final emissions tend to be overall underestimated. Satellite overpass times, cloud cover and fuel geometry all feed into active fire hotspot and radiative power product efficacy. Historically observations from AVHRR, GMS, GOES, MODIS and SEVIRI satellite sensors have been used to generate fire products. Current generation satellite sensors, including spatial resolution down to 375 m (e.g., with NPP-VIIRS) and a temporal resolution 10 minutes (e.g., from Himawari-8 and GOES 16) is possible when all available satellite data is used. This approach of multiple satellite sensor types is used to calculate emissions in near-real time with the FLAMBE and GFAS systems. Even so, differences between such databases in terms of fires processed and emissions can easily span an order of magnitude (Hyer et al., 2013). Even subtle differences in geolocation and fuel maps can result in substantial differences (Hyer and Reid, 2009).

- **Burnt Area**: The definition of burned areas works from temporal pairs satellite imageries, examining changes in an index derived from the solar reflective band measurements that show large differences when land changes from a vegetated to a “burnt" state. In this way, areas that have changed from “non burnt" to “burnt" between the date of the two satellite overpasses are highlighted, along with their best estimated date or burn. Shapefiles can be then be generated from this output, and cumulative area burnt can be derived from accumulations and filtering of this information. Burnt areas are therefore available after the event has ended (e.g., Barbosa et al. (1999)), however they provide an accurate estimation of how much dry matter has been burnt from which smoke emission can be calculated. The estimation of emissions from burnt areas has the advantages of being well established for a long time, being relatively close to in-situ methods employed locally by foresters on the ground (so that much validation and calibration has been performed in the past) and of being based on persistent burnt area signal that can be detected even after any observation gap, e.g. due to cloud cover. The spatial resolution is also relatively good, down to 250 m for global coverage. On the other hand, the burnt area observation is only possible after a burn so that true real-time applications cannot be realized. If an active fire satellite observation can only distinguish between “fire" and “no fire", e.g., due to its MIR channel saturating, this binary product is called a “hot spot" product. Sometimes, relatively simple assumptions are used to estimate burnt area from such products and emissions can subsequently be calculated as above. This has been done to correct for missing small fires in GFED (Randerson et al., 2012) or to calculate emission in real time, e.g. with FINN (Wiedinmyer et al., 2011). One of the best-known examples of burnt-area product is the GFED inventory Van der Werf et al. (2010).

- **Nighttime visible**: While active fire products have been the mainstay of fire emission estimates, historically some of the first satellite observations were associated with day/night imagers on the Operational Linescan System (OLS) on Defense Meteorological Satellite Program (DMSP; see Elvidge et al. (1996); review by Fuller (2000)). Increased availability of day/night band data from Suomi NPP VIIRS has renewed interest in fire detection through shortwave fire light (Miller et al. (2012); Polivka et al. (2016)).

- **Smoke detection**: Shortly after a fire is burning, its smoke plume can be detected in the atmosphere. Aerosol optical depth and carbon monoxide are relatively well observed by satellites, so that they can be used to infer the fire emissions using inverse methods. The advantage is that the atmospheric effect is relatively directly constrained for the observed species and that the effect of any unobserved fire is also included. On the other hand, this methodology cannot distinguish fire from other sources and has limited temporal and spatial resolution.

Combining the FRP and the burnt area has recently been shown to be a promising approach to derive quantitative estimates of fuel consumption.

3.3.2 Uncertainties in fire emission factors and BBA optical properties

Emissions of aerosols, and other pollutants, associated with open biomass burning are estimated using emission factors which convert between the mass of fuel consumed (derived from FRP or burnt area observations) and the species of interest via the carbon content of the fuel (e.g., Andreae and Merlet (2001); Akagi et al. (2011); Kaiser et al. (2012)). These emission factors are typically calculated using laboratory or field campaign measurements of smoke constituents which, while providing accurate measurements, may not be fully representative of all biomass burning and smoke conditions. In particular large uncertainties, and missing observations, persist in emission factors for different fuel types (e.g. peat), fire conditions (smouldering vs. flaming), and smoke processing scenarios (e.g. in the presence of clouds, day-time vs. night-time conditions) following, e.g., (Akagi et al., 2011). Increased and more extensive in situ measurements of different fire types would provide the data required to improve emission factors currently used in the operational models. Incorporating meteorological parameters, such as surface temperature, humidity and soil moisture, which could be done in NRT in the operational models will also be beneficial in adapting otherwise static emission factors to particular environmental conditions.

Peat fires are an important contributor to global carbon emissions, especially during ENSO related events in Indonesia (for example see the dedicated section in the BAMS State of the Climate 2015 (Benedetti et al., 2016) or Huijnen et al. (2016)). The signal from peat fires is relatively small and the proportionality to biomass burnt is less certain for these fires than for above-ground fires. Finally, the emission factors vary for individual fires so that estimates on a small scale have a limited accuracy. Observations that would help in better constraining the fire emission factors would be of great usefulness.

The uncertainties in emission estimates from smoke observations are still large due to variable and relatively poorly known optical properties of aerosols, poorly characterized errors of the used atmospheric chemistry and transport models, and noise in the satellite observations.

For burnt area products, uncertainties arise mostly from small fires remaining undetected in the burnt area observations and large uncertainties in the estimates of the rather variable input of available fuel load and combustion completeness. For peat fires in particular, the burn depth is not constrained with global observations.

3.3.3 Observations and data production for verification and assimilation

Beyond a regional increase in number and coverage of observations, near-real time access to site-observation data for assimilation and verification is an important point. Currently fire products from sensors on low orbit (MODIS, VIIRS) and geostationary satellites (SEVIRI, GOES, Himawari-8) are available. To estimate emissions, observation gaps may occur due to cloud cover or when satellite observations are not available, and the consistent merging of FRP from different satellites is still an open research topic, because their values are often very different and globally biased. However, combining the high temporal resolution of the geostationary products, which would greatly help in accounting for the usually strong diurnal cycle of fire emissions, and the higher precision and global reach of low Earth orbiting products is an important objective. Future satellite observations might help in reducing the discrepancy between low Earth orbiting and geostationary products.

To support the assessment of fire impacts, measurements of the combustion species (aerosols, reactive and greenhouse gases) are needed. There are several stations that can support verifications of haze forecast, but their number is very limited and some existing stations do not share data in a timely manner. There is also a network of ground-based observations, including Global Atmosphere Watch (GAW) stations and other global networks (e.g., AERONET). Lidar networks can also help with identifying the plume height.

Fire emissions occur most of the time in the Planetary Boundary Layer (PBL). However, for some large fires, estimated at roughly 15% of all fires, (Val Martin et al. (2010) and Sofiev et al. (2012)), fire emissions are released in the free troposphere above the PBL. In some extreme cases, fire emissions can even reach the Upper Troposphere – Lower Stratosphere region (Fromm et al., 2006). The height in the atmosphere at which this occurs is often referred to as the injection height. An observational dataset of injection heights exists through the MISR Plume Height Project (MPHP, Nelson et al. (2013)), based on a combination of MISR smoke aerosol and MODIS thermal anomaly products. This dataset has recently been updated and extended to produce the MPHP2 dataset. These observations have been very useful in calibrating and/or evaluating global biomass burning emissions injection height datasets (Sofiev et al. (2013)). Satellite products that can provide, in near-real time, an estimate of this injection height would greatly help in accurately forecasting large biomass burning events. Another factor of uncertainty, to a lesser extent, is also the shape of the vertical injection profile. In this case, profiling observations would be required.

3.4 User requirements for emissions: Desert mineral dust

For mineral dust, important source regions globally include the Sahara/Sahel, Southwest Asia/ Middle East, Taklimakan/Gobi deserts of China, Australia and the Southwest United States/adjacent Mexico. However throughout the world there are many individual sources such as in Patagonia, the Arctic plains, and countless dry or drying lake beds. Recent research has shown that current meteorological models struggle to realistically represent some key physical mechanisms of gust generation associated with dust emission (Knippertz and Todd, 2012). Surface wind speed is also poorly represented in many models and this induces errors both in dust emissions and subsequent transport (Menut et al., 2015). This is particularly true for northern Africa but many aspects apply to other source regions around the world, too. For example, many models create too much vertical mixing in the stable nighttime planetary boundary layer over arid areas, leading to an underestimation of nocturnal low-level jets and a too flat diurnal cycle in surface winds (Fiedler et al. (2013); Largeron et al. (2015); Roberts et al. (2017)). This is also partly related to an underestimation of turbulent dust emission during the day (Klose and Shao, 2012). Another substantial problem is the lack of dust generation related to cold pools (haboobs) associated with moist convection over the Sahel and Sahara (and many other desert areas), a process largely absent in models with parameterized convection (Marsham et al. (2011); Heinold et al. (2013); Pantillon et al. (2015, 2016)). This leads to even reanalyses missing the summertime maximum in dust generating winds in the central Sahara (Roberts et al., 2017). A too simplistic treatment of surface roughness in many models can also lead to significant differences in near-surface wind speed locally, particular in the semi-arid Sahel with its seasonal vegetation (Cowie et al., 2013; Kergoat et al., 2017).

It is challenging to improve model representation of dust generation due to an enormous lack of observations from key source regions. The logistically difficult and politically unstable Saharan and Middle East region has large areas void of any ground stations. What is required to better understand and specify the meteorology of dust production, is a much denser network of stations that observe standard meteorological parameters such as wind, temperature, humidity and pressure, ideally located in some of the main source regions. Given the large diurnal cycle and the short lifetime of some dust-raising mechanisms, particularly moist convection, an hourly or better time resolution would be desirable (Cowie et al., 2015; Bergametti et al., 2017). A first step in creating such a network was undertaken during the recent Fennec project (Hobby et al., 2013; Roberts et al., 2017). The lack of observations in combination with the difficult-to-represent meteorology also leads to substantial deviations between different analysis products, even on continental scales (Roberts et al., 2015), creating substantial differences in dust emission (e.g. Menut (2008)). Particularly the depth of the Saharan heat low, which is crucial for the large-scale circulation over northern Africa and thus a dominating factor for dust generation, can vary substantially between different analyses or model simulations with different resolution (Marsham et al., 2011). A much denser network of high-quality pressure observations is needed to better constrain models in this regard.

In addition, our knowledge of the amount and the size distribution of the emitted mineral dust particles is limited. This is to some extent due to uncertainties in the actual emission parameterization (e.g. Kok et al. (2014)), but also due to limited knowledge of relevant soil parameters that can vary markedly in space and time. To address this aspect, a network of ground stations is required that in addition to standard meteorology measures mineral dust emission, ideally including mass or number size distributions of emitted particles. Some such efforts were made during recent field campaigns such as Fennec (Marsham et al., 2013), the Bodélé Dust Experiment (BoDEx) (Washington et al., 2006) and the Japanese Australian Dust Experiment (JADE) (Ishizuka et al., 2008). Longer-term monitoring stations, however, are very rare, with the African Monsoon Multidisciplinary Analysis (AMMA) Sahelian Dust Transect being a notable exception (Marticorena et al., 2010; Bergametti et al., 2017). Worth mentioning are also the CV-DUST project (Pio et al., 2014) and the Cape Verde Atmospheric Observatory (CVAO) with its long term dust record (Fomba et al., 2014). An extension of such activities to more remote source areas would be highly desirable, especially one that accounted for the large particles. Further, knowledge about the mineralogy of the dust, i.e. the amount of soluble iron, is an important parameter to quantify the impact of dust emission on the biogeochemistry of the oceans (Mahowald et al., 2005).

Given the relative lack of in-situ data a continued reliance on remote sensing is anticipated in coming years, but a number of challenges remain. First, obscuration of dust by cloud (Kocha et al., 2013) is likely a problem that cannot be solved. Second, much summertime dust is emitted at night (Marsham et al., 2013) but most current products are day-time only, requiring better information from wavelength other than visible. Infrared products are being developed but still have biases related to atmospheric moisture (Banks et al., 2013). These would need to be further improved and provided in near-real time for data assimilation. Newly developed dust optical depth products from infrared high-spectral sensors such as Infrared Atmospheric Sounder Interferometer (IASI) are also promising (Peyridieu et al., 2013; Capelle et al., 2014). In addition, location of AERONET stations closer to source regions would allow evaluation of models and satellite retrievals near source (e.g., the short-term deployment during the Fennec field campaign Banks et al. (2013)), and retrievals from such observations should in future account for particles with diameters exceeding 30 μm (Ryder et al., 2013). Vertical profiles are also very important as discussed in Ansmann et al. (2017).

3.5 User requirements for emissions: Marine aerosol particles

Sea spray provides the largest mass flux of any aerosol type (Andreae and Rosenfeld, 2008) and sea salt aerosol dominates the total aerosol loading over the remote oceans (Haywood et al., 1999).

There are few long-term measurement sites of marine aerosol, all restricted to islands or coastal sites (e.g., MAN, http://aeronet.gsfc.nasa.gov/new_web/maritime_aerosol_network.html). The source of sea spray aerosol is strongly dependent upon environmental conditions, primarily the local surface wind speed, but also on wave state (Norris et al., 2013b), water temperature, salinity, and the presence of surfactants (De Leeuw et al., 2011). Biological material in the surface water can contribute to a significant organic component to the sea spray aerosol, increasingly so with decreasing particle size (De Leeuw et al., 2011). Most models, however, use simple source functions formulated in terms of wind speed only; the most widely used is that of Monahan et al. (1986), which is often applied well beyond the range of conditions from which it was derived and for which it is valid (Spada et al., 2013). Indeed, there appears to be a number of physical and biological effects that can strongly perturb the bubble/aerosol production relationship (Keene et al., 2017).

Extensive in situ measurement of aerosol particles within the marine atmospheric boundary layer is unlikely to be viable. Satellite remote sensing approaches offer the possibility of estimating both ambient aerosol loading and the source flux of marine aerosol. Passive measurement of reflected solar radiation can provide aerosol optical depth (Remer et al., 2005), and some information on both size and vertical distribution (Kokhanovsky, 2013). Active remote sensing can provide much better vertical resolution, and if multiple wavelengths are used, size distributions can be inferred. Both passive and active techniques suffer, however, from the fact that aerosol retrievals are only possible under cloud free conditions. Moreover, complicating matters is that there is more diversity in individual size measurements of sea spray than any other aerosol species (Reid et al., 2006).

The source of sea spray aerosol is breaking waves and the bursting of bubbles generated by them. Many source functions, including that of Monahan et al. (1986), scale a production flux of sea spray per unit area whitecap – integrated over its lifetime – by a whitecap fraction parameterized as a function of wind speed. There remains, however, an order of magnitude uncertainty in the parameterization of the whitecap fraction, and there is increasing evidence that neither the production of aerosol per unit area whitecap nor the lifetime of a whitecap are independent of the scale of wave breaking or other water properties (Norris et al. (2013a); Callaghan (2013); Spada et al. (2013), Salter et al. (2014, 2015)). Recent work on satellite retrievals of the whitecaps (Anguelova and Webster, 2006; Anguelova and Gaiser, 2011, 2013) shows significant promise as a means of providing this driving parameter for sea spray source functions, and implicitly accounting for the wide range of important controlling factors in addition to wind speed (Salisbury et al., 2013, 2014). It might, also, ultimately allow a source function to be specified directly in terms of the satellite measurements. While such an approach would provide near global coverage, the temporal sampling interval is dependent on satellite orbit.

The combination of satellite based estimate of both aerosol loading and source flux offer the optimum means of constraining operational model representation of marine aerosol. Future progress depends on improvements to, and validation of, the retrievals, and on improved estimates of the dependence of sea spray production on wave breaking and water properties. Measurements at very high wind speeds are also required to better constrain the parameterized source functions under extreme conditions, when sea spray production is greatest, for example during hurricanes or tropical storms.

3.6 User requirements for removal processes

Wet and dry deposition and sedimentation are important removal processes that control the predication the atmospheric aerosol distribution. However, the aerosol deposition fluxes themselves may become important NAAP forecast products, for example to forecast the soiling of solar panels.

The removal processes are modeled as a function of available meteorological variables describing boundary layer mixing. Wet deposition requires information about the occurrence of convection, precipitation and fog. Dry deposition modeling needs information about the state of the land surface and the vegetation, in particular for soluble aerosols. NAAP takes these meteorological variables from the underlying operational NWP models. It should be noted that improving the forecast of precipitation remains a major challenge for the NWP. Inaccuracies of the precipitation forecast directly influence the quality of the aerosol forecasts. Improving the surface information can be achieved by better linking NAAP to advanced land-surface modeling and by updating to the most recent land-use datasets.

Observations of wet deposition fluxes are available from acid deposition networks. These observations could be used to evaluate wet deposition of soluble aerosols such as sulphate, nitrates and ammonia. For this purpose, the observation need to be made available in a timely manner and at a temporal resolution suited for NAAP evaluation, which is often higher than the frequency (i.e. annual means) required for impact monitoring. Finally, the observed wet deposition flux are often strongly influenced by local processes, which makes it necessary to filter the observation in such a way that they are representative of the scale resolved by the NAAP models.

While some observation of wet deposition are made routinely, fewer observation of dry deposition observations are available. Uncertainties in deposition contribute substantially to the insufficient constraints in particular of the mineral dust mass budget in atmospheric mineral dust models. Currently, there are very few stations measuring dust deposition, both in the vicinity of and far from source regions (e.g., Bergametti and Fôret (2014)).

Measuring deposition fluxes is still a scientific challenge. Therefore the different sites often use different instruments and observational protocols (e.g., procedure to minimize contamination by local sources), which limits the comparability between observations. It is therefore desirable to enhance or develop standards for deposition measurements and to encourage continuous operation.

The Workshop on Measurement-Model Fusion for Global Total Atmospheric Deposition (MMF-GTAD), recently organized by the Global Atmosphere Watch Scientific Advisory Group for Total Atmospheric Deposition (SAG-TAD), explored the feasibility and methodology of producing, on a routine retrospective basis, global maps of atmospheric gas and aerosol concentrations as well as wet, dry and total deposition. In particular they reviewed the current state of global measurements (ground-based and satellite), chemical transport modeling (global and hemispheric), and measurement-model fusion/mapping techniques (see GAW report 234, at https://www.wmo.int/pages/prog/arep/gaw/documents).

4 Data assimilation for aerosol prediction

4.1 General concepts

An important aspect of aerosol prediction has been the development of data assimilation systems that include also gas and particulate phase species. Several global and regional models currently provide analysis of gases and aerosols. Currently five centers routinely assimilate aerosol data into their models, ECMWF/CAMS, FNMOC/NRL, GMAO, JMA, and UKMO. As an example among others, the CAMS system incorporates retrieved observations of ozone, CO, SO_2, NO_2 and AOD in its analysis, in order to provide initial conditions for the prediction of these species. As it is common in atmospheric composition, the assimilated data are products based on retrieval procedures. Bayesian, statistical or empirical methods are usually applied, depending on the complexity of the instruments and the observation characteristics. Direct assimilation of atmospheric clear-sky radiances in the UV, visible and near-infrared, where the aerosol signal is strongest, is being considered as a future step, which would allow a seamless assimilation of data from different satellite instruments. This has been shown possible in a study by (Weaver et al., 2007) but it has not been pursued in operational contexts as of now. There is a delicate trade off to achieve between the complexity and rapidity of the radiative transfer code in the shortwave. Complexity is required for simulating accurately clear-sky aerosol radiances in cases of low and high aerosol loads while rapidity is required in an operational context. Consideration of polarization might be necessary for the shorter wavelengths, thus further increasing the complexity and hence the computational cost of the radiative transfer calculations. The optimality of assimilating retrieved aerosol products versus radiances and the choice of a suitable algorithm or method for fast radiative transfer in the shortwave are thus still being debated. On the one hand direct radiance assimilation avoids the problem in the diversity between the model and the retrieval assumptions (aerosol type, refractive index, meteorological parameters, etc,), on the other hand the complexity of the observations might complicate or even prevent the implementation of radiance assimilation, especially for advanced sensors such as multi-angle instruments or polarimeters. In the end, the most pragmatic approach prevails in an operational context, hence the assimilation currently depends heavily on the availability of good quality retrieval products with some uncertainty estimates.

Emissions are not part of the analyzed fields but are specified either from established emission inventories (an extensive inter-comparison of various of these inventories is given in Granier et al. (2011)), from satellite observations as is the case for the emissions of biomass burning aerosols, CO and other species from wild fires (GFAS, Kaiser et al. (2012)), or computed in the model for some natural aerosol emissions (as sea spray or mineral dust). Estimation of emissions through data assimilation will be the next step for global models. This has already been successfully tried in regional models (e.g., Elbern et al. (2007)) and in off-line or online global models (i.e. Huneeus et al. (2012); Di Tomaso et al. (2017); Escribano et al. (2017)).

The most common approach is the adjustment of initial conditions in a manner similar to meteorological data assimilation used in Numerical Weather Prediction (NWP). Optimal interpolation, variational approaches (3D and 4D-Var), Ensemble Kalman Filter (EnKF) or hybrid techniques combining the advantages of both variational and EnKF techniques are all applicable and have been used at various operational centres in various flavors. Research is still ongoing for the optimal definition of the background error covariance matrices for aerosols, including errors deriving from the misspecification of the emissions. Hybrid 4D-Var/EnKF systems could be used to this end. Independently of the specific assimilation framework, assimilation is a key data-hungry application.

4.2 User requirements for data assimilation

In the past, the aerosol prediction and assimilation community had to use data that were being made available, and not necessarily aimed at the needs of this community. Aerosol products were often provided with climate applications in mind and made available as daily means or monthly averages. While the needs of the operational community are largely similar to those of the climate research community, the timeliness requirements are different. In recent years, with the advent of dedicated aerosol (and clouds) instruments, such as MODIS and CALIPSO, and the development of the model prediction of atmospheric composition, a new paradigm has been established. For example aerosol-related lidar missions such as EarthCARE and Aeolus, are now establishing best-effort near-real-time (NRT) data delivery, following the example of MODIS and CALIPSOs' expedited products. This has also been made possible by the fruitful collaboration between modelling community and data provider, in an effort to make an optimal use of the resources and provide the best service to the end-users.

At the moment, most aerosol assimilation systems rely on products such as AOD, rather than raw measurements such as satellite radiances. In the case of lidar measurements, aerosol backscatter, attenuated backscatter or extinction are all candidate variables for assimilation. However, the tendency in the future may be towards the use of satellite radiances, either raw or aggregated and possibly cloud-cleared, for consistency with the current approach in NWP. This represents a challenge for both the model developers and the data providers and might also involve joint development of observation operators.

Some general recommendations related to data assimilation observational requirements are outlined below.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-42
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 27 February 2018

4.2.1 Timeliness

Observations of key variables have to be timely. In particular, especially for aerosol prediction and air-quality applications, the data to be fed into the assimilation system need to be in near-real time (NRT, i.e. available within 6 hours) and have an associated time-stamp.

4.2.2 Uncertainty

Regarding the user requirements on uncertainties for assimilation applications, two main points should be highlighted:

1. Observation errors on the assimilated product have to be provided at the pixel or retrieval level. Statistical error models can help to understand the general accuracy of the data product, but are not so useful for data assimilation where the observations are considered pixel-by-pixel. Wherever possible error covariances should also be provided, which include correlations of errors between different aerosols products from a given sensor, correlations of errors in time (especially for retrievals from geostationary satellites), and correlations of errors in space (e.g., due to the similarity in surface properties or viewing geometries). Additionally, other information is deemed necessary for the correct assimilation of the observations, such as averaging kernels for chemical species. Moreover, retrieval errors should be required to stay below a certain threshold in order to make the cut for assimilation.

2. Biases should be quantified and, where possible, filtered out before data provision for assimilation. Even sophisticated assimilation systems with online bias correction struggle with aerosol observations as there is limited redundancy at the moment and no single satellite sensor can be used as an absolute reference as they all suffer from biases. Ground-based lidars and sun-photometers are being currently investigated to provide a bias-free anchoring for satellite data or as a dataset to be assimilated on their own right (Rubin et al., 2017). This approach shows a lot of promise, provided that the calibration of the ground-based instruments is monitored and possible sources of biases in the processing of the data are removed.

Provided that random and systematic errors are provided, the assimilation can “cope” with large errors, given the fact that errors (both in the background and in the observations) appear as weighting factors. If the error in the observation is large compared to the difference between the model and the observation (departure), then that particular observation will have only a minor influence on the analysis. This is particularly true for unbiased random errors. For systematic errors this is not true. Unless biases can be removed, if the differences between the model and the observations are too large the assimilation cannot cope and the observation in question is usually rejected on the assumption of perfect model, which is often made in, for example, variational assimilation. Generally the analysis is the result of a statistical compromise between error assumptions on the model background and on the observations. There is limited tolerance of biases, but the main assumption behind the most common estimators for data assimilation is that they are linear and unbiased.

4.2.3 Spatial resolution and sampling

The requirement on spatial resolution of the observations needed for assimilation is quite relaxed due to the fact that current global assimilation for operational aerosol prediction cannot afford to run very high-resolution analysis. For this reason, even data with coarse spatial resolution (100 km) can be beneficial. However, in most cases, current satellite-based sensors have a much better spatial resolution down to few kilometres for passive sensors and few hundred meters for active sensors (depending on the application). Spatial sampling is possibly more important than resolution for assimilation. It has been shown that assimilation of an instrument with large spatial sampling (wide swath) such as MODIS is more beneficial than assimilation of high accurate measurements from a passive sensor with a narrow swath. However, using observations from narrow-swath instrument adds value to the analysis. From the point of view of the ground-based networks, the density is an important factor. Vertically resolved observations are also very important, even if the spatial resolution is not very high, since they provide information regarding the vertical structure of the aerosol field which is completely missing in the integrated AOD measurements which are provided currently. Lidar backscatter and extinction profiles provide the necessary vertical information and the challenge remains to integrate this information with that provided by the passive sensors. This entails both improving the modelling and the retrieval aspects.

4.2.4 Temporal resolution

The issue of temporal resolution is similar to the spatial resolution. In principle high-temporally resolved data are beneficial to the analysis, but issues connected to large data volume may arise. This is particularly true for datasets coming from geostationary satellites which have to be heavily thinned. This is obviously only a technical limitation which might not be applicable across the range of assimilation system. For ground-based instruments, similar considerations can be made, although data volume might not be as high.

4.2.5 Speciation

The problem of constraining the aerosol species in the model has become more important with user demand of products related to single aerosol types. Providing forecast of AOD constrained by observations is not enough as detailed speciated information on dust, biomass burning and anthropogenic aerosol particles is needed for several applications. For example, a large portion of CAMS users is interested in dust forecasts for energy-related applications. From the point of view of the NWP, having robust aerosol climatologies to use in the radiation scheme is a necessity. However, it is not only total AOD that is of interest but the extinction connected to the single species since their radiative impact depends on refractive index which is in turn a function of chemical composition. Recently, the CAMS climatology was compared to the Tegen et al. (1997) climatology as far as impact on NWP scores at ECMWF (Bozzo et al., 2017). Although, on one hand both the total AOD and the dust AOD in the CAMS climatology appear more realistic if compared with independent measurements, on the other hand the impact in the model is mostly positive in summer. In winter, the impact is neutral or slightly negative due to the lack of absorbing aerosols in the CAMS climatology in the winter hemisphere particularly over Europe and North America. This is a model shortcoming which has to be addressed by improving the description of the emissions and secondary production of organic aerosols. The most recent version of the CAMS model includes for example a parameterization for Secondary Organic Aerosols (Samuel Rémy, private communication). Data assimilation however can partially help to constrain the problem if appropriate speciated information can be included. At the moment the main observation is total AOD which is used to constrain either total AOD itself or total aerosol mixing ratio. In some models the control variables in the assimilation are the individual species but there is no information on speciation contained in the AOD: the same value of AOD can be obtained by different combinations of the AODs of the individual aerosol species. This implies that any information on speciation comes from the model itself, regardless of the degree of sophistication of the assimilation.

Rather than assimilating total AOD, it seems more desirable to assimilate coarse-mode AOD (e.g., dust and sea-salt) and fine-mode AOD (e.g., pollution and wildfires) independently. However, if both fine and coarse mode AODs are retrieved using the same measurements, the correlation of their errors would have to be provided. Absorption AOD (AAOD) would also be a good parameter to constrain the absorbing aerosols in the model, particularly for NWP application as this parameter controls the amount of heating induced by aerosols in the atmosphere. This effect can sometimes counteract the surface cooling that non-absorbing aerosols have (Chylek and Wong, 1995). Wherever direct speciation measurements are possible, those would be the best suited to be used to correct model prediction of a given aerosol species. This could be measurements derived from a (relatively dense) network of ground-based instruments or from satellites.

Recent improvements in lidar retrievals are also indicating the possibility to discriminate speciation information from these profiling information, at least for certain aerosol species such as dust and volcanic aerosols. For dust, more specifically, a few simple meteorological parameters could be also pointed out referring back to section 2.

1. Surface pressure observations from northern Africa to better constrain pressure gradients and therefore winds, which are not directly assimilated into models.

2. Surface temperature and dew point help to better constrain soil moisture in the top soil layer in most data assimilation systems. This can be particularly important for dust from semi-arid areas like the Sahel as well as East Asian semi-arid areas, where seasonal soil moisture and vegetation can be a major factor for uncertainties.

4.2.6 Resilience

Several data sources are needed to ensure resilience of the system and a wealth of observation-based information. Currently most centres rely on satellite data for the analysis of aerosols. The next generation of satellite measurements is designed to provide more information on the horizontal and vertical distribution of atmospheric particulate but current efforts often focus on trace gases, while aerosols products are often considered secondary. It is important to consider that some satellites currently providing vital information for aerosol assimilation are coming to the the end of their lifetime (for example MODIS). it will be therefore crucial that there are concerted efforts to replace such instruments and insure continuation of data provision and long-term consistency of the records. Frequent instrumental changes may cause problems for data uptake and recalibration of biases corrections, impinging as well on the quality of the forecast products. Efforts are also under way to use ground-based and aircraft measurements.

4.2.7 Format and accessibility

Finally, observations have to be available in a format that is easily accessible, and should also be as compatible as possible with model fields. For example, it could be more useful to report fine and coarse mode AOD at a reference wavelength (as it is more relevant to modal schemes in global modeling) rather than or in addition to the Angström exponent (AE) (O'Neill et al., 2003). However, errors on AODs at multiple wavelengths are correlated, while errors in AE retrievals tend to be only weakly correlated with those in AOD, making AE a possibly more attractive variable for assimilation. This is actually still a matter of debate in the retrieval and assimilation communities.

It is also recommended that mechanisms are put in place for easy data transfer, especially for heavy-duty users.

5 Evaluation of aerosol forecasting models

5.1 General concepts

Evaluation is an important step in operational prediction. NWP has well-established evaluation protocols of prediction products, whereas similar procedures for aerosol forecasting are still being defined (Reid et al., 2011). The poorly developed verification system for NAAP compared to NWP, together with the lack of standardized evaluation processes and suitable observations limit the advancement of operational aerosol prediction. One major difference between NWP and NAAP verification is that NWP often relies heavily on verification of a forecast system against its own (or another) analysis. This approach seems much less suitable for NAAP where the observational constraint in the analysis is much weaker.

For operational forecasting purposes, we distinguish between two different evaluations: model evaluation that is conducted as soon as observations of the forecast period are made available and one where the model's performance in simulating a given event or longer time period (e.g., seasonal or annual cycle) is examined in depth. The latter is related to benchmark testing. Operational evaluation, sometimes referred to as verification, is generally part of the operational forecasting process and is therefore done on a regular basis in near real-time, whereas benchmark evaluation can made any time after the forecast period, and observations that were not available for the near real-time evaluation can be included. Furthermore, while the operational evaluation allows to quantify the confidence and predictive accuracy of the model products and quickly identify problems which may arise in the forecast, benchmark testing identifies weaknesses of individual models and provides an assessment of model performance and uncertainties. This is in turn a useful information for the forecast users.

It is not within the scope of this paper to list and describe the requirements for an extensive model evaluation associated to model developments. This could involve various different aspects of the aerosol life cycle such as aerosol-cloud interactions, heterogeneous chemistry, removal processes, etc. Each one of these aspects would require a large and specific set of observations. In the present section, we will focus on the evaluation conducted as part of the implementation of an operational forecast. For operational purposes, it is important that these observations are delivered timely and on a regular basis, to ensure the possibility of a routine evaluation. As pointed out in 2, in addition to aerosol measurements it is also important to include meteorological and chemical observations in the model evaluation process to complement and understand the resulting aerosol predicted fields. Moreover, taking into account that there are operational forecasting systems with data assimilation, it is important to include independent observational datasets (not used during the data assimilation process) in the model evaluation.

Satellite remote sensing is the most convenient tool for providing global aerosol spatial and temporal distributions. However, it is difficult to discriminate the satellite aerosol signal from surface reflectance, which is the reason why algorithms based only on the visible spectral region often fail over bright surfaces (Hsu et al., 2004). In the last years a number of new advanced sensors (like MODIS, OMI, CALIOP, IASI, MISR or SEVIRI) have been launched on board polar or geostationary satellites. Usually, polar satellites (such as Aqua, Terra or CALIPSO) are at relatively low altitudes (between 500 and 800 km), covering a global domain at high spatial resolution. However, the polar sensors provide few measurements per day over the same point. On the other hand, geostationary satellites are situated at a set point over the Equator, at a 36000-km height, and provide measurements over a given disk. For instance, Meteosat Second Generation (MSG), which hosts the SEVIRI radiometer, is an essential tool for NRT monitoring in Europe and Africa.

Since the atmospheric residence time of aerosol particles is relatively short and the footprint area of a single station may be limited, there is a need for observation networks with sufficient density of stations. A description of the current and future needs for the observing system has been provided in Laj et al. (2009). Clearly, all analyses are pointing to the need for improving geographical coverage of measuring stations. Point measurements are biased towards populated areas as in Europe and United States (see Figure 1). Data collected from commercial aircraft can provide invaluable observations for model evaluation (e.g., In-service Aircraft for a Global Observing System, IAGOS; http://www.iagos.org/). At the moment, however, this is not established for operational aerosol applications.

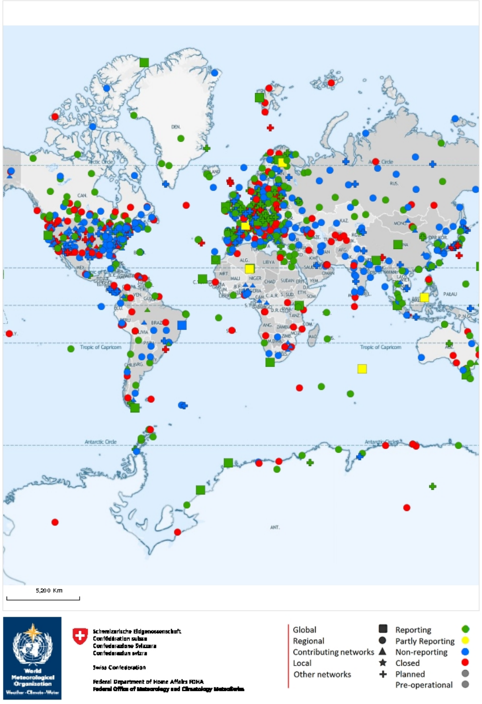

Figure 1. Map of surface stations currently included in GAWSIS.

Various ground-based observational systems are in operation to monitor aerosol properties in the atmosphere (GAW report 2016) that can be either policy, science-driven or both. Their organizational structure may vary. Among the main contributors to the aerosol observing system are:

- Near-Surface concentration measurements: the European Monitoring and Evaluation Programme (EMEP; http://www.emep.int), the Interagency Monitoring of Protected Visual Environments (IMPROVE) program (http://vista.cira.colostate.edu/improve/), the African Monsoon Multidisciplinary Analysis (AMMA) Sahelian Dust Transect (Marticorena et al., 2010);

- Aerosol variables at near-surface provided by the European Research Infrastructure ACTRIS (Aerosol, Cloud and Trace Gases Research infrastructure, http://actris.eu) and the NOAA ESRL's Global Monitoring Division with its specific program on aerosol monitoring (https://www.esrl.noaa.gov/gmd/aero/);

- Vertical profiles : the European Aerosol Research LIdar NETwork (EARLINET; http://www.earlinet.org), which is part of ACTRIS, the Asian Dust and Aerosol Lidar Observation Network (AD-Net; http://www-lidar.nies.go.jp/AD-Net/), the Latin America Lidar Network (ALINE; www.aline.org), the NASA Micro-Pulse Lidar Network (MPLNET, https://mplnet.gsfc.nasa.gov/);

- Aerosol optical depth: the global Aerosol RObotic NETwork (AERONET; http://aeronet.gsfc.nasa.gov) (Holben et al., 1998), the Global Atmospheric Watch Precision Filter Radiometer (GAW-PFR) Network (Wehrli, 2008), the sky radiometer network (SKYNET; http://www-lidar.nies.go.jp/skynet/) (Takamura et al., 2004). Details on homogeneity of AOD from different networks can be found in Kim et al. (2008).

- Wet deposition: EMEP, the Acid Deposition Monitoring Network in East Asia (EANET, http://www.eanet.asia/) and the National Atmospheric Deposition Program (NADP, http://nadp.sws.uiuc.edu/) and the Canadian Air and Precipitation Monitoring Network (CAPMoN)in North America.

All these networks (except AERONET, AMMA and EMEP) are GAW contributing networks. They consists of 32 stations which cover different types of regions documenting variability of aerosol properties: clean and polluted continental, marine, arctic, dust, biomass burning, and free troposphere. While global stations are expected to measure as many of the key variables as possible, the approximately 300 GAW Regional stations generally carry out a smaller set of observations.

The most widely used network is AERONET providing measurements of AOD as well as Aerosol Size Distribution (ASD) with over 600 sites around the world. Most stations report in NRT and products are used at several centres for both routine and retrospective validation.

While recognizing the current efforts, there is still the need to secure long-term funding for ground-based stations and to further develop infrastructure and data protocols in order to fully support forecasting aerosol activities. Much effort has also been dedicated to standardize protocols and formats to ensure quality assurance, traceability and data quality of aerosol observations from both ground-based and space-based sensors but it is still a challenge.

Most of the routine measurements (e.g., hourly or daily basis) are conducted as part of regional networks/infrastructures and their usage for operational evaluation is limited to the configuration of the model used in the forecast. Depending on the model resolution, global models do not always capture the spatial variability of the individual stations, particularly for mountain sites and urban sites where the influence of unresolved topography and/or local emission sources are dominant factors for the aerosol distribution. Station data are sometimes selected if representative of background conditions or they may be aggregated. For that reason, for model evaluation, it is mandatory to provide additional information on the observation site with a correct classification based on its spatial representation (regional or global) and its localization (environment types and emission types). Otherwise, detailed information on the model individual aerosol components has become more important in the last years. Speciated information on dust, biomass burning and anthropogenic aerosol particles is needed for several socio-economic applications (e.g., solar energy and air quality). However, the presence of different types of aerosols mixed in the measurement points should introduce errors in the comparison between individual aerosol model outputs and observations.

5.2 User requirements for operational evaluation

Operational evaluation is specific for models used operationally. It involves operational online verification of model output, plausibility checks and quality control. As in the case of data assimilation (see Section 4.2), high-temporally resolved data are needed. The operational evaluation is an assessment of how the forecast behaves relative to observations that are near-real-time (i.e., available within 24 to 48 hours since the forecast run), allowing the modeling group and the end users to have a quick overview of the quality of the forecast. Note that the timeliness requirement is less stringent than for assimilation. At present the most used product to evaluate aerosol model outputs in NRT are surface and/or satellite based atmospheric column integrated variables such as AOD (at a reference wavelength of 550 nm). Only recently have products like aerosol size distribution, aerosol scattering or absorption coefficients become available in NRT from a limited number of stations. However, the ability of a model to reproduce AOD may not always be a good indicator of its performance to reproduce surface concentration or vertical aerosol distribution (Huneeus et al., 2016), even though these model variables are clearly inter-connected. Therefore and in the absence of emission and deposition routine observations, model evaluation should combine atmospheric column integrated variables with vertical profiles (extinction coefficient at a reference wavelength at 550 nm to provide information about the height and thickness of the aerosol layer), and surface measurements (such as PM10, PM2.5 and PM1). They also provide an evaluation of the aerosol size distribution on surface level. For the atmospheric column, the Ångström exponent (which provides aerosol size information) and the separation of AOD into fine-mode and coarse mode contributions can be used to evaluate the aerosol size distribution.

Additionally, since data sets of weather surface records have better spatial and temporal coverage, observations of horizontal visibility included in meteorological reports are used as an alternative way to monitor aerosol events in near-real-time (NRT) and to qualitatively evaluate the aerosol forecasts. In addition, key meteorological variables as surface winds (linked to emission of natural aerosols) and precipitation (linked to wet deposition) should be considered.

5.3 User requirements for benchmark testing

Benchmark testing examines individual processes and input drivers that may affect model performance and requires detailed atmospheric measurements that are not, typically, routinely available and can provide better quality control. In addition to those variables considered in the operational evaluation, benchmark testing is expected to include as many of the key variables as possible. Comprehensive measurements of aerosol size distributions, chemical composition, and optical properties are needed. Such observations should ideally be collocated with detailed meteorological information and vertical distribution (e.g., lidars and radiosondes).

Routine, long-term measurements of aerosol size distributions, chemical composition and optical properties in operational ground-based networks are urgently needed for model evaluation. These measurements should include the following: mass concentrations of chemical components (soot, organics, ammonia, sulphate, nitrate, mineral dust and sea salt), number concentrations (of PM_1, $PM_2.5$, PM_10), and size distribution (if possible resolved by chemical species; e.g. separately for $< 1\ \mu$m and $> 1\ \mu$m).

Evaluating whether relevant emission and feedback processes are treated accurately by a model is challenging, although data assimilation can provide valuable information (Pope et al., 2016). In addition to key meteorological parameters associated with aerosol emission (e.g., surface winds and soil moisture), the effects of aerosols on radiation and clouds, for example, depend on the physical and chemical properties of the aerosols.

Evaluating direct and semi-direct aerosol effects on aerosol absorption properties requires aerosol optical properties such as aerosol absorption optical depth (AAOD), particle depolarization (relative to aerosol speciation), altitude distribution (relative to clouds), radiation observations such as solar irradiance (downward and net shortwave radiation, downward longwave radiation and outgoing longwave radiation) and solar surface albedo. Evaluating indirect aerosol effects on clouds and precipitation is even more challenging and it would require additional detailed observations of cloud properties such as cloud optical depth, cloud droplet number concentrations or cloud top height and thickness (used to evaluate aerosol and deep/shallow convective cloud interactions).

For benchmark testing, there is also a need for co-located and simultaneous meteorology and chemistry measurements at locations carefully selected to ensure spatial representativeness. To fully understand processes, more sites with co-located observations of visibility, cloud, radiation, vertical profiles of temperature, relative humidity as well as winds and aerosol properties would be highly desirable. Precipitation and deposition observations are also extremely relevant for benchmarking. Innovative designs for global measurement systems (existing technological platforms such as commercial aircraft, cell phones, cars, etc.) should be further exploited. Such a task should fit the mandate of international organizations such as WMO and EUMETNET (see GAW report 226 on Coupled Chemistry-Meteorology/Climate Modelling, available from WMO).

5.4 Format and accessibility

As in the case of data assimilation (see section 4), observations used in the model evaluation have to compatible with the model output fields. In this sense, it would be desirable to work on the establishment of formats and common protocols for data harmonization and exchange. This is the main objective of the Data Centers. At present, there are six GAW World Data Centers (WDCs) each responsible for archiving one or more GAW measurement parameters or measurement types. They are operated and maintained by their individual host institutions. They collect, document and archive atmospheric measurements and the associated metadata from measurement stations world-wide and make these data freely available to the scientific community. In some cases, GAW WDCs also provide additional products including data analyses, maps of data distributions, and data summaries. However, each GAW WDCs is treating their databases independently even if different communities are providing the same aerosol parameter. This fact can introduce some discrepancies in the definition of one parameter, creating problems for the model to observation comparisons.

6 Conclusions

Numerical atmospheric aerosol prediction is at a crossroads. It has experienced quick progress in the recent years due to the availability of aerosol models, aerosol satellite observations, data assimilation techniques and the knowhow of numerical weather prediction. This paper takes stock on past achievements and reflects on how further progress can be made with a focus on user requirements for aerosol measurements in the context of operational prediction. Requirements are discussed in relation to modeling, assimilation and evaluation, and concern resolution, sampling, accuracy and timeliness of the observations. However it was felt that no hard-line requirements can be set up in terms of goal, threshold and breakthrough values given the relative youth of NAAP. Rather this study aims at developing the needs of a new community and establish scientific criteria based on which those values can be defined at a later stage. At this moment, there is a more pressing need to recognize that measurements of aerosol particle properties are not only a “nice-to-have” element in operational and research observing ground-based networks and space-borne platforms, but they are instead an important and necessary part of the Global Observing System.

Further improvements to NAAP will likely follow several directions:

- Better representation of aerosol processes. This will require to pitch the right level of complexity (especially in terms of chemical speciation), get the best possible meteorological information from NWP, and the relevant aerosol measurements to calibrate and evaluate aerosol parametrisations.

- Improved data assimilation, both in terms of techniques and choice of aerosol variables to be assimilated. Key questions for the future are whether there is a benefit to move from assimilating AOD to assimilating clear-sky radiances in the shortwave spectrum and how to make the best possible use of vertical profiles from lidar observations.

- Better aerosol data fueled by a stronger integration of NAAP with aerosol data providers and clear presentation of user requirements. NAAP ought to better consider the issue of aerosol speciation and aerosol size distribution in aerosol modelling, data assimilation and verification.

Concerning aerosol requirements, we recommend the following stepwise approach. The community should start with a better quantification of requirements for total mass, chemical speciation and the size distribution at the surface with the aim to improve emissions and boundary layer processes. Second, similar requirements will also be required in the free troposphere, in order to better constrain long-range transport, sedimentation and interaction with radiation. Third, having this information, the next step would be to understand better how the various data streams complement each other (or not) in the context of global operational aerosol prediction in order to assess which additional data is expected to improve most the aerosol forecast.

Acknowledgements. The authors gratefully acknowledge Oksana Tarasova (WMO) for supporting this initiative and encouraging the Scientific Advisory Group on Aerosols to take the lead in writing this paper. Many thanks to: Silvina Carou, Markus Fiebig, Sang-Woo Kim, John Liggio, Sarah Lu, Olga Mayol-Bracero, Hu Min, John Ogren, Gelsomina Pappalardo, Andrea Petzold, Nobuo Sugimoto and Judd Welton. Mark Parrington is gratefully acknowledged for his contribution to the fire emission section. Thanks also to Zak Kipling for his useful comments to the final draft of the manuscript. Angela Benedetti has received funding from the H2020 Aerosols, Clouds, and Trace gases Research InfraStructure (ACTRIS2, Grant Number 654109). Sara Basart acknowledges the AXA Research Fund for funding aerosol research at the Barcelona Supercomputing Center through the AXA Chair on Sand and Dust Storms. InDust (COST Action CA16202) is also gratefully acknowledged. This is NOAA PMEL contribution number 4739.

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