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Recent developments in monitoring and modelling airborne pollen, a review

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Abstract

Public awareness of the rising importance of allergies and other respiratory diseases has led to increased scientific effort to accurately and rapidly monitor and predict pollen, fungal spores and other bioaerosols in our atmosphere. An important driving force for the increased social and scientific concern is the realization that climate change will increasingly have an impact on worldwide bioaerosol distributions and subsequent human health. In this review we examine new developments in monitoring of atmospheric pollen as well as observation and sourceorientated modelling techniques. The results of a Scopus[®] search for scientific publications conducted with the terms "Pollen allergy" and "Pollen forecast" included in the title, abstract or keywords show that the number of such articles published has increased year on year. The 12 most important allergenic pollen taxa in Europe as defined by COST Action ES0603 were ranked in terms of the most 'popular' for model-based forecasting and for forecasting method used. Betula, Poaceae and Ambrosia are the most forecast taxa. Traditional regression and phenological models (including temperature sum and chilling models) are the most used modelling methods, but it is notable that there are a large number of new modelling techniques being explored. In particular, it appears that Machine Learning techniques have become more popular and led to better results than more traditional observation-orientated models such as regression and time series analyses.

Keywords: Aerobiology, Phenology, Aeroallergen, Pollen forecasting, Real-time pollen monitoring networks, Machine Learning

Public awareness of the rising importance of allergies and other respiratory diseases has led to increased scientific effort to accurately and rapidly monitor and predict pollen, fungal spores and other bioaerosols in our atmosphere. An important driving force for the increased social and scientific concern is the realization that climate change will increasingly have an impact on worldwide bioaerosol distributions and subsequent human health (Beggs et al. 2017).

This is symbolised by the results of a Scopus[®] (Elsevier B.V.) search for scientific publications we conducted with the terms "Pollen allergy" and "Pollen forecast" included in the title, abstract or keywords (Source: Scopus, December 2019). The results obtained show that the number of such articles published has increased year on year (Figure 1). In the 1970s, there were less than 150 papers on pollen allergy and only the occasional paper on pollen forecasting published each year. By the end of the 2010s, this increased to about 600 pollen allergy papers (maximum 618 in 2017) and 12 pollen forecasting papers (maximum 16 in 2014) published annually.

This increased interest in pollen allergy contributed to the creation of several European Cooperation in Science and Technology Actions (COST), these were for the "Assessment of production, release, distribution and health impact of allergenic pollen in Europe (ES0603 -EUPOL)", the "Sustainable Management of Ambrosia artemisiifolia in Europe (FA1203 -SMARTER)", and the current network for examining "New approaches in detection of pathogens and aeroallergens (CA18226)". One of the main outputs from COST Action ES0603 was the influential book entitled "Allergenic Pollen", edited by Mikhail Sofiev, Karl-Christian Bergmann, which has more than 10,000 downloads since it was published in 2013. The book examined pollen sources (Skjøth et al. 2013), the onset, course and intensity of the pollen season (Dahl et al. 2013), monitoring, modelling and forecasting of the pollen season (Scheifinger et al. 2013), airborne pollen transport (Sofiev et al. 2013a), the impact of pollen (de Weger et al. 2013) and

the presentation and dissemination of pollen information (Karatzas et al. 2013). However, since then there have been a number of notable advancements in the field of aerobiology, particularly the development of devices capable of automated real-time monitoring (e.g. Oteros et al. (2015) and Crouzy et al. (2016)) and the continued development of numerical forecast models (Sofiev et al. 2015).

This review paper came from the Pollen Monitoring and ModELling (POMMEL) project that aims to produce a reliable and fully operational pollen forecast system for Ireland (https://www.pommel.ie). The focus of this review is to examine the current state-of-the-art in pollen monitoring and forecasting for the 12 most allergenic taxa as defined by COST Action ES0603 (i.e. *Alnus, Ambrosia, Artemisia, Betula,* Chenopodiaceae (Amaranthaceae), *Corylus,* Cupressaceae, *Olea, Platanus,* Poaceae, *Quercus,* Urticaceae (*Urtica, Parietaria*) (Skjøth et al. 2013)), with particular emphasis on developments since the publication of the pivotal work "*Allergenic Pollen*".

Advances in bioaerosol monitoring

Before we can talk about recent developments in modelling and forecasting airborne pollen concentrations, we need to discuss recent advances in bioaerosol monitoring as one is intimately linked to the other. Today, the vast majority of monitoring networks still use technology that dates back to the middle of the last century (Buters et al. 2018). For instance, many aerobiological networks are based on volumetric spore traps of the Hirst (1952) design, which were developed some 70 years ago. This technology is not without its problems and workers have reported differences between Hirst type traps located only a few meters apart (Tormo Molina et al. 2013) and errors in determining the flow rate (Oteros et al. 2017a). The Hirst type trap is by no means the only sampler used in networks and another widely used volumetric/impaction technique is the Rotorod (Grinnell et al., 1961); a rotating mechanism that is said to have a sampling efficiency of ~85% (Noll 1970).

Hirst-type sampling protocols require at least one full day before the drum can be changed and the sample processed. Although this period is quite often extended to 7-days for logistical and financial reasons. Subsequent analysis of the pollen-covered tapes by light microscopy requires high levels of skill to accurately count and properly identify the particles collected. This is a slow process and, as a result, only a small sub-sample of individual microscope slides are examined and the overall count is an extrapolation (Comtois et al. 1999; Šikoparija et al. 2011). The data also relies on the skill of the operator and so there have been concerted efforts of late to evaluate data quality (Galán et al. 2014; Sikoparija et al. 2017a; Smith et al. 2019) and standardise methods (Galán et al. 2014; Galán et al. 2017). Having said this, however, there are reasons why such equipment has been in operation for so long. Hirst type traps are relatively inexpensive to purchase and operate. The technique is also robust and performs well outdoors (Beggs et al. 2017). Furthermore, long-term datasets are now available with wide geographical distributions that can be used for examining changes in plant phenology and distribution (Ziska et al. 2011; Ziello et al. 2012; Smith et al. 2014; Sikoparija et al. 2017b).

One of the main problems (or greatest weaknesses) that pollen monitoring networks face is the time delay between sampling, analysis and eventual communication of the findings to the general public and health care professionals. As a consequence it is now recognised that aerobiological studies should focus on creating reliable, real-time monitoring networks. A comprehensive review of the real-time sensing of bioaerosols has been published by Huffman et al. (2020) and several of these methods are discussed below.

The KH-3000-01 samples a portion of air containing particles by use of a laser beam to irradiate the air flow (Kawashima et al. 2007). It makes real-time pollen monitoring possible

because the light-scattering particle data are processed by a computer instantaneously. The device has been developed by Yamatronics Corporation (Japan) and employs one light source and two light receptors, which are able to discriminate Japanese cedar or cypress pollen from other particles. It is able to do so by comparing scattered light intensity and degree of polarization. Its low-cost makes it an attractive proposition for piloting a real-time pollen monitoring network.

Instrumentation based on the fluorescence spectroscopy of biological particles has recently been developed to provide rapid, on-line, real-time counting (if not yet identification) of bioaerosols. For instance, the Waveband Integrated Bioaerosol Sensor (WIBS-4 - now superseded by the WIBS-NEO) has been described in a number of publications (O'Connor et al. 2014; O'Connor et al. 2015; Fennelly et al. 2018) and it has been successfully deployed in the field resulting in strong correlations with traditional volumetric techniques (e.g. R² >0.9) (O'Connor et al. 2014; Calvo et al. 2018).

The PA-300, and its successor the Rapid E, is also available (Crouzy et al. 2016; Huffman et al. 2020). The instrument uses the measurement of fluorescence signals as a simple diagnostic to distinguish between airborne particles that emit light and those that do not. The technique offers great potential for the rapid determination of the fluorescence properties of target samples, in addition to being non-destructive and reagent free.

Auto-fluorescence is a known property of biological particles because their structural components (e.g. tryptophan, flavonoids, chitin, lignin, "sporopollenin" and secondary metabolites such as phenols and terpenoids) have been shown to absorb and emit light over a range of visible and UV wavelengths (Roshchina 2008; Pöhlker et al. 2012). Hence the WIBS and the Plair instruments, in particular, have been applied to the detection of many types of

bioaerosols in both the laboratory and field settings (Healy et al. 2014; Crouzy et al. 2016; Feeney et al. 2018).

In Europe, two automatic real-time pollen networks are now being established. The first is in Bavaria, Germany, and uses BAA500 automatic image recognition technology (Oteros et al. 2015). Whilst in Switzerland, MeteoSwiss is establishing a real-time pollen monitoring network upon the Swisens Poleno air-flow cytometry system (Sauvageat et al. 2020). Currently real-time devices such as the BAA500 and Swisens Poleno surpass the budgets available to most national pollen service providers. They are also relatively untested. In fact, only four European countries (France, Germany, Luxemburg and Switzerland) are monitoring with automatic samplers and not in all their locations (Buters et al. 2018). There are also two locations in the USA, and a further 120 in Japan where only a few taxa are of allergenic interest to the population (Kawashima et al. 2007).

Alternative molecular biology methods have been developed for examining aeroallergens. For example, pollen and fungal spore allergens sampled from the atmosphere can be quantified using enzyme-linked immunosorbent assays (ELISA) and reagents are available in kit form. Studies using such techniques have shown that the presence of pollen allergens in the atmosphere do not always correlate with the occurrence of airborne pollen grains (Buters et al. 2012; Buters et al. 2013; Galan et al. 2013), which could impact on the information provided to allergy sufferers and strategies for managing their symptoms. The use of DNA metabarcoding can also provide information on the ecology of the atmosphere (Brennan et al. 2019). Nonetheless, molecular techniques are still a long way from being able to monitor the atmosphere on a routine basis.

Modelling and forecasting the pollen season

It is hoped that the cost of automatic real-time monitoring devices will decrease in the near future due to increasing commercial production and expertise. This would allow much faster sampling and analytical processing to occur, thereby lengthening forecast horizons and improving forecasts. Furthermore, measurements with high temporal resolution will also increase our knowledge of atmospheric processes (Šikoparija et al. 2018b; Chappuis et al. 2020). Here we discuss the two main approaches for modelling and forecasting airborne pollen as described in Scheifinger (2013); observation-orientated and source based models.

Observation-orientated models

Observation-orientated models are based on our environmental knowledge of a certain location. For instance, average pollen concentrations from the same day in previous years can turn pollen calendars into forecasting tools (Šikoparija et al. 2018a). In observation-orientated mathematical and statistical models the dependent variables are phenological observations or airborne pollen concentrations that can be predicted using one or more independent variable (e.g. meteorological data). The main disadvantage of this approach is that it is site-specific, which can lead to difficulties in extrapolating to other locations if the model is not adjusted appropriately. With this in mind, it should be noted that Oteros et al. (2019a) successfully used a combination of linear regressions and Kriging interpolation to spatially model pollen concentrations for unmonitored areas in Bavaria, Germany.

Traditional methods include regression and time series analyses and process-based phenological models. On the other hand, so-called Machine Learning (or Deep Learning) is a new technique that has been successfully integrated into many aspects of data mining over the

last few years. Indeed, it is seen as an analytical solution in many different disciplines and applications such as self-drive vehicles or image recognition (Zhang et al. 2018).

Regression analysis

Linear regression (two quantitative variables, one of them dependent Y, and other independent, X, explained by a straight line) or multiple regression analyses (one quantitative variable, dependent Y and more than two quantitative independent variables X₁...X_k being related in a linear or non-linear way) work by predicting future values for one dependent parameter, such as pollen, by means of one or more parameters including their historical observations (Kinnear & Gray 1999). Paul Comtois (1998) entertainingly stated that "because of its predictive values, the establishment of a regression is often pompously called model building, even in reality it concerns only the establishment (and testing) of a slope (b) and of a correlation coefficient (r)".

Nevertheless, due to the relative ease of construction regression models remain popular in aerobiological studies and a number of papers have been published in recent years that include simple linear regression analysis (Piotrowska-Weryszko 2013b; Bonini et al. 2015; García-Mozo et al. 2015; Frenguelli et al. 2016; Picornell et al. 2019b) as well as polynomial and multiple regression analysis (Sabariego et al. 2012; Ocaña-Peinado et al. 2013; Oteros et al. 2013a; de Weger et al. 2014; Donders et al. 2014; Novara et al. 2016; Ritenberga et al. 2016; Tseng et al. 2018). Approaches include the more traditional stepwise or backwards elimination multiple regressions (Sicard et al. 2012; Aboulaich et al. 2013; Myszkowska 2013; Howard & Levetin 2014; Malkiewicz et al. 2014; Zhang et al. 2015; Murray & Galan 2016; Janati et al. 2017; Robichaud & Comtois 2017; Galera et al. 2018; Volkova & Severova 2019), which are by far the most frequently used, as well as logistic or 'logic' regressions (Escabias et al. 2013; Myszkowska 2014b; Myszkowska & Majewska 2014; Katz & Batterman 2019), Partial Least Squares (Brighetti et al. 2014; Oteros et al. 2014; Aguilera et al. 2015a; Bogawski et al. 2019b; Lara et al. 2019) and Generalized Linear Model (Charalampopoulos et al. 2018).

Regression analysis continues to be used to model daily average airborne pollen concentrations (Janati et al. 2017) in addition to different characteristics of the pollen season including start date (Myszkowska 2014a; Zhang et al. 2015; Novara et al. 2016), peak day (Myszkowska 2013), duration (Zhang & Huang 2015) and intensity (Oteros et al. 2013a; Bonini et al. 2015). Regression models have been constructed for different pollen types including Alnus (Piotrowska-Weryszko 2013b; Myszkowska 2014b; Novara et al. 2016), Ambrosia (Howard & Levetin 2014; Zhang et al. 2015), Artemisia (Piotrowska-Weryszko 2013a; Zhang et al. 2015), Betula (Myszkowska 2013; Inatsu et al. 2014; Zhang et al. 2015; Robichaud & Comtois 2017; Tseng et al. 2018; Bogawski et al. 2019b), Corylus (Myszkowska 2014b; Frenguelli et al. 2016; Novara et al. 2016), Cupressaceae (Sabariego et al. 2012; Ocaña-Peinado et al. 2013; Charalampopoulos et al. 2018; Picornell et al. 2019b), Olea (Sicard et al. 2012; Oteros et al. 2013a; García-Mozo et al. 2014; Frenguelli et al. 2016; Charalampopoulos et al. 2018; Picornell et al. 2019b), Platanus (Frenguelli et al. 2016; Charalampopoulos et al. 2018; Picornell et al. 2019b), Poaceae (Piotrowska 2012; Aboulaich et al. 2013; de Weger et al. 2014; Zhang et al. 2015; Janati et al. 2017; Picornell et al. 2019b), Quercus (Fernández-Llamazares et al. 2014; Jato et al. 2014; Frenguelli et al. 2016; Picornell et al. 2019b) and Urticaceae (Picornell et al. 2019b).

Despite their convenience, regression techniques do not take into account seasonal variations in daily temperatures during the year or the associated increases in pollen concentrations in Spring and Summer. Time-dependence is also an important factor when considering the potential impact of external influences, like environmental change, on long-

term datasets of plant phenology. Such factors can be included in models using time series analysis. These are a sequence of observations for which a variable Y is sorted by sampling date (with a certain level of correlation amongst them) as a function of time (t).

Time-series analysis

Time series analysis is a technique used in aerobiology for obtaining insight from the underlying relationships observed in the data, by studying longer datasets with an aim to forecast the possible future behaviour of the parameter Y (usually, but not always, sometimes it can be used to reconstruct past time series). The time series are associated with various controlling components of variability, the two most important are defined below (Aznarte M et al. 2007; Scheifinger et al. 2013; García-Mozo et al. 2014; Rojo et al. 2017):

General trend (G). — The predominant behaviour that Y shows from the start to the end of the series. It is expected to be unidirectional and smooth, clearly showing future expectations, be it an increase, a stationary trend or a decrease.

Seasonality (S). — Small fluctuations along the timeline associated with natural cycles of less than one year in length. They are ordinarily repeated every year in a recognised way.

However, in spite of having two well identified components, sometimes the behaviours of some natural parameters are much more complex.

unknown cycles (C). — They may also contribute. Often, these are cycles observed in more than one-year period and could provide explanations for variability, especially in changing climatic conditions (e.g. North Atlantic Oscillation or El Niño–Southern Oscillation). These variables are obviously very complex and so sometimes they are simply included as part of the general trend.

Nonetheless, the complete time series approach clearly requires one further component of variability before an accurate, overall model can be constructed:

Random components (R). — Possible fluctuations without a known seasonality or pattern, which make it impossible to accurately predict future behaviour.

Therefore the full general time series predictive model can be summarised as:

$$Y(t) = G(t) + S(t) + C(t) + R(t)$$

Time series analyses are intended to separate (decompose) different patterns with the goal of isolating all the disturbances provoked in the time series datasets by ordinary seasonal behaviour (S) such as weather parameters. However, in addition to ill-defined random components, residual noise (R) can be present as a consequence of inter-annual variability or unknown factors.

As a result of this decomposition, the general trend (G) is shown in a more accessible way to be interpreted, and we can avoid that other parameters mask the true pattern. In this sense, seasonal-trend decomposition procedure based on LOcally wEighted Scatterplot Smoothing (LOESS smoothing) has been applied with more interest in aerobiological studies lately (Rojo et al. 2017). For example, Rojo et al. (2017) found the decomposition of time series to be suitable for analysing the phenological and meteorological factors influencing airborne Poaceae pollen concentrations, including seasonal effects of species with different pollination periods. García-Mozo et al. (2014) also used the decomposition procedure based on Loess (STL), and an ARIMA (Auto Regressive Integrated Moving Average) model to examine changes in the flowering pattern of *Olea* in Córdoba (Spain) in relation to global climate change. Although

Aguilera et al. (2015b) were not able to verify these findings even though they used a similar method for decomposing seasonal time series.

Time series analyses have been performed for many years for different pollen types (e.g. see Scheifinger et al. (2013) and references therein). Recently, time series analyses have been used to model airborne concentrations of *Alnus* (Siniscalco et al. 2015; Nowosad et al. 2016), *Ambrosia* (Puc & Wolski 2013; Siniscalco et al. 2015), *Artemisia* (Puc & Wolski 2013; Siniscalco et al. 2015), *Betula* (Nowosad et al. 2016), *Corylus* (Nowosad et al. 2016), Cupressaceae (Silva-Palacios et al. 2016), *Olea* (García-Mozo et al. 2014; Aguilera et al. 2015); Fernández-Rodríguez et al. 2016c), *Platanus* (Siniscalco et al. 2015), Poaceae (Puc & Wolski 2013; Brighetti et al. 2014; Tassan-Mazzocco et al. 2015; Fernández-Rodríguez et al. 2016a; Rojo et al. 2017; Fernández-Rodríguez et al. 2018), *Quercus* (Fernández-Rodríguez et al. 2016b), and Urticaceae (Tassan-Mazzocco et al. 2015; Valencia et al. 2019).

Process-based phenological models

Phenology is the timing of naturally recurring seasonal events in animals and plants, such as migration and flowering. Phenological phases are influenced by meteorological parameters, particularly temperature, and as such can be used as a proxy for climate change (e.g. Thackeray et al. (2010), Ziello et al. (2012) and Dahl et al., (2013) and references therein). Phenological observations of plants can be separated into different stages of the flowering process, such as anthers closed, anthers open and full flowering (Lukasiewicz 1984). On the other hand, characteristics of the airborne pollen season like start date, peak day and duration, which are also phenological phenomena, can be defined using different techniques (Jato et al. 2006). Phenological records can complement aerobiological data (Tormo et al. 2011) but temporal mismatches have been identified between flowering phenophases and the appearance of

pollen in the air (Estrella et al. 2006). With long-distance transport of pollen being an important source of uncertainty (Linkosalo et al. 2010; Sofiev et al. 2013b).

Phenological models determine the recorded dates of phenological phases as a function of environmental factors (e.g. chilling temperature, forcing temperature, photoperiod, and water availability) and are important components of numerical atmospheric transport models. For example, Sofiev et al. (2013b) described the emission module for the SILAM numerical model of birch pollen emission and dispersion (System for Integrated modeLling of Atmospheric coMposition, http://silam.fmi.fi). The output of this pollen emissions module is described as a 'release flux' of pollen grains (Sofiev et al. 2006; Sofiev et al. 2013b). The pollen emission module proposed by the authors follows the principle of a double-threshold thermal time phenological model described by Linkosalo et al. (2010) that can describe the whole period of flowering from beginning to end (Linkosalo et al. 2010; Sofiev et al. 2013b). It has been noted that the simplest thermal time models perform better than more complicated approaches (Linkosalo et al. 2008; Sofiev et al. 2013b). Indeed, Pauling et al. (2014) tested parameterizations of temperature sum models for 12 pollen stations in Switzerland. The authors examined a simple thermal model that relies solely on forcing temperatures and a sequential model that also used chilling temperatures, and concluded that the addition of chilling did not substantially improve the statistical skill of the models (Pauling et al. 2014).

Several studies have been published in recent years that have included forcing temperatures (Pauling et al. 2014; Achmakh et al. 2015; Linkosalo et al. 2017; Picornell et al. 2019a) and chilling (Siniscalco et al. 2015; Novara et al. 2016). Most phenological models were constructed for spring flowering trees of the Betulaceae family namely *Alnus* (Pauling et al. 2014; Siniscalco et al. 2015; Novara et al. 2016; Linkosalo et al. 2017), *Betula* (Pauling et al.

2014) and *Corylus* (Pauling et al. 2014; Novara et al. 2016), but also for *Olea* (Achmakh et al. 2015) and Poaceae (Pauling et al. 2014).

Machine Learning

Since more traditional models often poorly depict the intricate relationships between various factors influencing airborne pollen concentrations (Nowosad 2016), more complex methods such as machine learning are often employed to overcome these limitations. Machine learning (ML) is the application of computational techniques that mimic characteristics of biological information processing systems for data modelling (Recknagel 2001). As previously discussed by Scheifinger et al., (2013), the use of these advanced techniques is on the rise due to their ability to out-perform deterministic models. In recent years this trend has continued with more and more studies utilizing ML methods to forecast pollen concentrations.

A number of ML techniques are available with Artificial Neural Networks (ANNs), Support Vector Machines (SVMs) and Random Forests (RFs) being some of the most widely discussed for pollen forecasting. However, other ML techniques have been studied with less popularity, such as Least Absolute Shrinkage and Selection Operator (LASSO) (Liu et al. 2017), plus Cubist algorithms and Multivariate Adaptive Regression Splines (MARS) (Nowosad et al. 2018).

In recent years, Artificial Neural Networks have become particularly popular in modelling the non-linear behaviour of pollen (Astray et al. 2016). ANNs are composed of individual neurons in multiple layers, mimicking the biological neural system and is able to learn from complex, noisy, incomplete data to model non-linear relationships (Valencia et al. 2019). Deep learning occurs when there is an intrinsic learning in the neural network and the number of hidden layers is greater than two. Deep learning ANNs contain a number of epochs (iterations), which improve the learning capability of the ANN and thus the performance (Zewdie et al. 2019b). The application of ANNs have assessed a variety of aspects relating to the modelling and forecasting of pollen. For example, Multilayer Perceptron ANNs (MLPs) have been used to predict the daily *Ambrosia* concentration over different cities up to 7 days ahead (Csépe et al. 2014) and in the construction of a *Ambrosia* pollen alarm system in the Pannonian biogeographical region (Csépe et al. 2020). The use of ANNs to model aerobiological data is not new (Sánchez-Mesa et al. 2002; Arca et al. 2004; Sánchez Mesa et al. 2005; Aznarte M et al. 2007; Rodríguez-Rajo et al. 2010) and more recent studies have been employed to model the relationship between pollen taxa such as *Ambrosia* (Csépe et al. 2014; Csépe et al. 2019; Zewdie et al. 2019a), *Betula* (Puc 2012), *Quercus* (González-Naharro et al. 2019), *Olea* (Oteros et al. 2013b; Iglesias-Otero et al. 2015) with various meteorological, phenological and remote sensing parameters. Current applications of ANNs have included the adaption of existing models to improve performance in emission models, e.g. Burki et al. (2019) improved the efficiency of the COSMO-ART dispersion prediction model by combining ANN functionality.

Support Vector Machines also express complex non-linear relationships by learning but do so by using Vapnik-Chervonenkis dimensional theories (Du et al. 2017). Unlike, ANNs and RFs, very few studies have exclusively employed SVMs for pollen forecasting and are generally compared alongside other modelling methods. SVMs in the past have been used in developing models for the prediction of flowering periods (Bogawski et al. 2019b) as well as forecasting daily pollen concentrations (Zhao et al. 2018; Zewdie et al. 2019a). In these studies, SVMs tend to perform better than deterministic regression models but are often out performed by ANNs and RFs, perhaps equating to the lack of independent SVM studies.

Random Forests differ slightly from the other methods in that it is an ensemble technique and involves constructing a series of decision trees. This method generates a number

of regression trees relating to the sample data and then combines them by averaging (Navares & Aznarte 2020). This method has the added capability of determining the most important variables contributing to a model (Zewdie et al. 2019a) and as such have been used in several studies to identify the most important variables for improving ML models (Navares & Aznarte 2017; Navares & Aznarte 2020). Aside from this, RFs have been used to develop prediction models for high concentrations of different pollen types, such as *Alnus, Betula* and *Corylus* (Nowosad 2016; Nowosad et al. 2016) and Poaceae (Navares & Aznarte 2017). However, the performance of RFs and the other ML methods can produce poor predictions if the range of training data is insufficient (Bogawski et al. 2019b). As such, comparisons between different ML methods are often conducted to determine the best performing model.

Model performance is strongly dependent on the data provided, particularly when it comes to model training. Overfitting is often observed if the model corresponds too strictly to training data to the extent that it learns the noise and fluctuations, which in turn impacts negatively on the model's ability to adapt to other data (Zanotti et al. 2019). The extent of overfitting depends on the model type, the predictor variables as well as the correlation between those variables and the output (Nowosad et al. 2018). Every model can overfit but ANNs are notorious for illustrating this behaviour (Zanotti et al. 2019). Cross-validation is commonly used to prevent overfitting and involves producing a series of miniature training sets from the original training data to essentially tune the model. A variety of cross validation, and k-fold cross-validation (Nowosad et al. 2018). Pruning is another well documented algorithm used to limit the effect of model overfitting and is commonly applied to RFs (Singh et al. 2013; Csépe et al. 2014). Alternative methods to correct overfitting include bootstrap

aggregation/bagging (Navares & Aznarte 2020; Soundiran et al. 2019), gradient boosting (Dai et al. 2019) and Bayesian Regularization (Balram et al. 2019; Zanotti et al. 2019).

To determine the best machine learning method for the task, many studies have opted to compare various ML techniques. In fact, the majority of ML studies for predicting pollen concentrations cover a selection of modelling methods. For instance, Csépe et al. (2014) compared a series of MLP and tree based algorithms for the prediction of Ambrosia pollen concentrations and Nowosad et al. (2018) compared nine different modelling techniques including both linear and non-linear models for Alnus, Betula and Corylus pollen concentrations. Additionally, Bogawski et al. (2019b) predicted the onset of the Betula flowering period by comparing five types of model, including: SVMs, RFs, partial least squares, ordinary least squares and linear regression. Similarly, Zewdie et al. (2019b) employed RF, extreme gradient boosting, deep neural networks and linear Bayes ridge models to forecast the airborne Ambrosia concentration over Tulsa. These studies illustrate the superior performance of ML methods over less sophisticated models. RFs (Liu et al. 2017; Nowosad et al. 2018; Bogawski et al. 2019b) and ANNs (Zewdie et al. 2019b), especially MLPs (Csépe et al. 2014) have performed particularly well in past studies and their applications will likely be further explored in the future.

Pollen production, release and dispersal are affected by a plethora of factors and, in order to improve the development of future ML models, authors have suggested the incorporation of additional parameters to improve model performance such as spatial location, the distribution of potential sources, phenological data, past pollen concentrations, chemical air pollutants and land use (Csépe et al. 2014; Nowosad 2016; Nowosad et al. 2018). One leading concern is the tendency of ML models to underestimate pollen concentrations during days of unusually high concentration. These errors can arise for several reasons, one being as a result

of mass transport from distant sources. Therefore, focus should be placed on the incorporation of forward and back trajectories into ML models (Zewdie et al. 2019b). Since one ML method cannot accurately predict all pollen types in various locations efficiently, future work is likely to include additional comparisons and combinations of various advanced modelling techniques (Voukantsis et al. 2010). There has also been a notable progression from point source meteorological data to weather radar. Recent studies have illustrated the benefits of utilizing gridded meteorological data opposed to station data, which is confined to its point scale and may not fully represent the study area (Nowosad 2016; Zewdie et al. 2019c). Moreover, weather radar allows for the prediction of pollen concentrations over larger spatial scales and is increasing in popularity (Zewdie et al. 2019b; Zewdie et al. 2019c).

Overall, Machine Learning models represent some of the most promising pollen modelling methods and are likely to see large developments in the coming years with the increasing prevalence of aeroallergens and availability of remote sensing databases, including those coming from the Sentinel 2 project (Ottosen et al. 2020).

Source-orientated models

Source-orientated pollen emission and transport models quantify and forecast temporal and spatial distributions of near surface airborne pollen levels (Verstraeten et al. 2019). These models are based on Chemistry Transport Models (CTM's) that have been extended to deal with the atmospheric dispersal of pollen and, unlike observation-orientated models, require knowledge of source conditions and calculations of diffusion (Scheifinger et al. (2013), Sofiev et al. (2013a) and references therein). Sofiev et al. (2015) provides an overview of the seven models in the European MACC modelling consortium: CHIMERE, EMEP, EURAD-IM, LOTOS-EUROS, MATCH, MOCAGE, and SILAM. In addition, other source-orientated numerical forecast

models able to simulate the dispersion of pollen include COSMO-ART (Vogel et al. 2008; Vogel et al. 2009; Zink et al. 2012; Zink et al. 2017) and ENVIRO-HIRLAM (Mahura et al. 2009) in Europe, and CMAQ-pollen (Efstathiou et al. 2011) in the USA. Furthermore, the WRF-CHEM model (Weather Research and Forecasting model coupled to Chemistry), developed by the American National Oceanic and Atmospheric Administration, has been used in the UK (Skjøth et al. 2015c). Recent studies examining the use of source-orientated models include the following pollen types: *Alnus* (Prank et al. 2016), *Ambrosia* (Zink et al. 2012; Prank et al. 2013; Liu et al. 2016; Prank et al. 2015; Prank et al. 2016; Sofiev 2017; Pauling et al. 2020), *Olea* (Hernandez-Ceballos et al. 2014; Zhang et al. 2014; Prank et al. 2016; Sofiev 2017), and *Quercus* (Zhang et al. 2014), Poaceae (Prank et al. 2016; Sofiev 2017), and *Quercus* (Zhang et al. 2014; Jeon et al. 2018).

A major cause of uncertainty in source-orientated models is data of pollen emission sources (Sofiev et al. 2006; Verstraeten et al. 2019). For instance, Verstraeten et al. (2019) increased correlations between SILAM modelled and observed time series of daily average *Betula* pollen concentrations using a combination of an updated source inventory of *Betula* trees derived from forest inventory data, MODIS vegetation photosynthesis data (GPP), and updated start and end dates of airborne *Betula* pollen seasons. This so called 'bottom-up' approach of preparing source inventories using forest inventories was advocated by Skjøth et al. (2008a) and Pauling et al. (2012). Conversely, a 'top-down' approach combining land use data, annual pollen indices and local knowledge of species distribution has been successfully used for mapping the invasive and highly allergenic *Ambrosia artemisiifolia* (Sikoparija et al. 2009; Skjøth et al. 2010; Thibaudon et al. 2014; Karrer et al. 2015; Bonini et al. 2018). Zink et al. (2017) showed that the *Ambrosia* pollen inventory for France published by Thibaudon et al.

(2014), produced using this top-down approach, resulted in the best agreement between COSMO-ART simulated and observed airborne *Ambrosia* pollen concentrations.

The identification of potential source regions of airborne pollen has been a productive area of research in aerobiology in recent years, and examples include the Concentric Ring Method for Olea (Rojo et al. 2016) and Quercus (Oteros et al. 2017b). Air mass trajectory analysis has traditionally been used to identify potential sources of pollen such as Ambrosia (Stach et al. 2007; Sikoparija et al. 2009; Skjøth et al. 2009; Kasprzyk et al. 2011) and Betula (Hjelmroos 1991; Hjelmroos 1992; Skjøth et al. 2008b; Skjøth et al. 2009). The analysis of air masses continues to be an accepted method for investigating the paths taken by airborne pollen. Ambrosia (Sommer et al. 2015; Grewling et al. 2016; Bilińska et al. 2017) and Betula (Skjøth et al. 2015a; Skjøth et al. 2015b; Izquierdo et al. 2017; Bogawski et al. 2019a) remain popular subjects, but target species have been expanded to include Alnus (Skjøth et al. 2015a; Skjøth et al. 2015b; Bilińska et al. 2019), Artemisia (Qin et al. 2019), Olea (Fernández-Rodríguez et al. 2014; Hernandez-Ceballos et al. 2014), and Quercus (Skjøth et al. 2015a; Maya-Manzano et al. 2016). The use of remote sensing techniques such as LiDAR (Katz & Batterman 2019; Pecero-Casimiro et al. 2019) and interpolation techniques like Kriging (Oteros et al. 2019a; Pecero-Casimiro et al. 2019) represent the present state-of-the-art.

Discussion and Conclusions

The 12 most important allergenic pollen taxa in Europe as defined by COST Action ES0603 were ranked by Karatzas et al. (2013) in terms of the most 'popular' for model-based forecasting and for forecasting method used. An update of these results (2012-2019) shows that *Betula* and Poaceae are still the most forecast taxa. Although *Betula* has overtaken Poaceae for first place and the invasive *Ambrosia* has replaced *Olea* in third (Table I). A broad range of pollen

forecasting methods are now available for researchers. Regression and phenological models (including temperature sum and chilling models) remain the most used methods (Table II), but there has been a sea change and it is notable that there are a number of new modelling techniques being explored (16% of the total) (Table III). This includes Bayesian statistics (Zhang et al. 2013; Jochner-Oette et al. 2019), which are difficult to pigeonhole along with other methods discussed. The fact that the most popular forecasting goal is now numerical modelling reflects this change (Table I). In particular, it appears that Machine Learning techniques are becoming more accepted and have led to better results than more traditional observationorientated models such as regression and time series analyses.

Forecasting atmospheric concentrations of pollen is not generally considered to be excessively expensive, because the computational resources required are not exceptionally demanding and the necessary data for processing are not difficult to obtain. Improvements in monitoring are likely to drive further innovations in forecasting as real-time information becomes available for data hungry models. Although it is essential we continue to monitor environmental change by extending long-term datasets, e.g. by maintaining some sites that use traditional Hirst type traps, as seen in the Bavarian model (Oteros et al. 2019b). Finally, it is important that fungal spores are also studied and modelled. This is because of their allergic impact and because they can penetrate deep into our respiratory system due to their small size (PM_{2.5} range for some of the most important). There is presently less literature concerned with modelling airborne fungal spores, compared to pollen, but this will also change in the future as real-time methods become more prevalent.

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Pollen	Daily	Season	Peak	Season	Season	Phenology	Long Term	Total	Total from
Таха	Values	Start	Day	Duration	Severity	(1 Month		2012-2019	Karatsas et al.
									(2013)
Alnus	1	6	1	1	2	1	2	14	23
Ambrosia	7	6	2	2	3	4	1	25	3
Artemisia	-	3	1	1	-	1	2	8	1
Betula	4	11	2	5	7	4	2	35	40
Chenopodiaceae*	-	-	-	-	-	-	-	0	0
Corylus	1	5	1	1	1	2	2	13	11
Cupressaceae	3	2	1	2	2	-	1	11	4
Olea	2	5	4	2	4	5	2	24	24
Platanus	1	3	1	1	-	-	-	6	9
Poaceae	9	6	2	3	3	2	5	30	42
Quercus	3	3	1	2	1	2	2	14	19
Urticaceae	1	1	1	1	-	-	-	4	3
Total	32	51	17	21	23	21	19	184	179

Table I. The forecasting goal for the ranked 12 most allergenic pollen taxa in Europe for model-based forecasting. An update of results from a literature analysis of pollen forecasting models from 2012 to 2019. Data from Karatsas et al. (2013) included for comparison.

The table reports on the forecasting goal for each one of 12 pollen taxa in Europe (note that more than one target may be mentioned per taxa).

NB*Amaranthaceae

Table II. The most 'popular' forecasting methods, models and factors for modelling used for the 12 most allergenic pollen taxa in Europe as listed by Karatsas et al. (2013) and updated in this study (literature analysis of pollen forecasting models from 2012 to 2019). Data from Karatsas et al. (2013) included for comparison.

Methods used in Forecast models	Alnus	Artemisia	Ambrosia	Betula	Chenopodiaceae*	Corylus	Cupressaceae	Olea	Platanus	Poaceae	Quercus	Urticaceae	Total	Total from Karatsas et al. (2013)
Agromet. Coefficient			1							1			2	1
Artificial Neural Networks	1		5	3		1		4		3	1	2	20	6
Atmospheric transport model	2	2	3	7				4	2	1	3		24	2
Chilling	11	1	1	6		4	1	3	2		5		34	29
Dispersion Model	1	1	5	6				3	2	2	1		21	1
Linear discriminant analysis								1		1			2	1
Multiple component analysis Non-linear logistic regression				1									1	1
model Phenological/Climatological	2		1	1		1			1	1		1	8	4
methods	5	1	4	10		3	1	8	1	3	4		40	32
Poisson regression	1									1			2	2
Polynomic regression										2			2	1
Regression	13	4	5	30		11	7	19	7	32	13	5	146	69
Simulated anealing	1	1	1	1				1	1	1	1		8	3
Temperature SUM	11		1	10		5	2	10	3	2	7		51	45
Time series analysis	4	1	3	12		4	1	3	1	9	1	2	41	8
Total	52	11	30	87	0	29	12	56	20	59	36	10	402	205

The table reports on the forecasting goal for each one of 12 pollen taxa in Europe (note that more than one target may be mentioned per taxa).

NB* Amaranthaceae

Table III. The most popular new modelling techniques organized by the studied pollen taxa and not previously mentioned in Karatsas et al. (2013). An update of results from a literature analysis of pollen forecasting models from 2012 to 2019.

Methods used in Forecast models	Alnus	Artemisia	Ambrosia	Betula	Chenopodiaceae*	Corylus	Cupressaceae	Olea	Platanus	Poaceae	Quercus	Urticaceae	Total
Bayesian Statistics			1										1
Classification and Regression Trees (CART)								3					3
Cubist	1			1		1							3
Emission Model			2	1				1	1		2		7
functional logit regression								1		1			2
Gradient boosting			1										1
Gumbel's distribution		1											1
LASSO	1		1	1		1							4
Linear Mixed Models Multinomial logistic regression								1					1
analysis	1			1		1				1			4
Multivariate Adaptive													
Regression Splines (MARS)	1			1		1							3
Partial Least Squares	1			3		1		4		1			10
Random Forests (RF)	3		5	4		3				2			17
Support Vector machines	1			3		1							5
Total	9	1	10	15	0	9	0	10	1	5	2	0	62

The table reports on the forecasting goal for each one of 12 pollen taxa in Europe (note that more than one target may be mentioned per taxa). NB* Amaranthaceae