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Ozone modelling and mapping for risk assessment: An overview of different approaches for human and ecosystems health

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ABSTRACT

Tropospheric ozone (O_3) is one of the most concernedair pollutants dueto its widespread impacts on land vegetated ecosystems and human health. Ozone is also the third greenhouse gas for radiative forcing. Consequently, it should be carefully and continuously monitored to estimate its potential adverse impacts especially inthose regions where concentrations are high. Continuous large-scale O_3 concentrations measurement is crucial but may be unfeasible because of economic and practical limitations; therefore, quantifying the real impact of O_3over large areas is currently an open challenge. Thus, one of the final objectives of O_3 modelling is to reproduce maps of continuous concentrations (both spatially and temporally) and risk assessment for human and ecosystem health. We here reviewedthe most relevant approaches used for O_3 modelling and mapping starting from the simplest geo-statistical approaches andincreasing in complexity up to simulations embedded into the global/regional circulation models and pro and cons of each mode are highlighted. The analysis showed that a simpler approach (mostly statistical models) is suitable for mappingO_3concentrationsat the local scale, where enough O_3concentration data are available. The associated error in mapping can be reduced by using more complex methodologies, based on co-variables. The models available at the regional or global level are used depending on the needed resolution and the domain where they are applied to. Increasing the resolution corresponds to an increase in the prediction but only up to a certain limit. However, with any approach, the ensemble models should be preferred.

1. Introduction

Because of its widespread presence in urban and rural environments, air pollution is a serious threat forany life-form and especially for animaland plant health (Lelieveld et al., 2015; Sicard et al., 2016a). The increasing pollutant emissions inmany regions of the worldare perceived as the second biggest environmental concern by citizens, after climate change (EEA, 2019). This perception results in an increasing level of attention for media and citizens. This growing public engagement, which includes ongoing citizen science initiatives supporting air quality monitoring (EEA, 2019) and initiativesto increase public awareness and behavioural changes around air pollution challenges, hasled to growing expectations for measures aiming at preventing severe risk for human health. Tropospheric ozone (O_3) is one of the most important atmospheric pollutants in terms of detrimental effects on human (Cohen et al., 2017; Sicard et al., 2021a) and ecosystems health (Li et al., 2018;

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Feng et al., 2019), as well as on biodiversity conservation (Agathokleous et al., 2020).

Ground-level $O_3$is a secondary pollutant formed by its precursors (nitrogen oxides, NO_x; Volatile Organic Compounds, VOCs; and methane) in the presence of sunlight; its concentrations are influenced by anthropogenic and natural emissions, chemical, physical, and biological processes (Lamarque et al., 2013). Local and remote pollution sources, atmospheric chemical processes, long-range transport (Huang et al., 2018; Jonson et al., 2018) and stratospheric influx (Knowland et al., 2017)all affecttropospheric O_3 concentrations. In addition to anthropogenic sources, natural processes such as El Niño-Southern Oscillation (ENSO) conditions influence tropospheric O_3 production (Rowlinson et al., 2019). Increments in surface O_3 concentrations contribute to changes in air quality (Sicard et al., 2020; Sicard, 2021), human health (Cohen et al., 2017), forest growth and vitality (Proietti et al., 2016; Feng et al., 2019) and agricultural productivity (Van Dingenen et al., 2009). Tropospheric O_3contributes to increasing global warming both directly, due to its radiative effects (Checa-Garcia et al., 2018), and indirectly, by affecting photosynthesis and ultimately reducing the land carbon sink capacity (Sitch et al., 2008).

Air quality conditions require, as the first and essential step, reliableestimatesof air pollutants concentrations. Different O_3 measurement techniqueshave beenrecently reviewed bySaitanis et al. (2020), showingthatair quality monitoring stations cannot cover all the territory, due to economic and technical limitations. Indeed, monitoring networks are often spatially heterogeneous with limited geographic coverage, for instance in China (Sicard et al., 2021b) or in the United States (Bravo et al., 2012). Identifying the environmental factors that influence air pollutantslevels over these areas is thus necessary to establish new locations for extending the network representativeness. Due to the impossibility to directly measure O_3 concentrations in all areas, the modelling approach is a useful and suitable tool to indirectly obtain information on O_3 concentrations over large regions. Researchers and technicians use models as integrative tools for risk assessment of O_3 pollution and policy evaluation in several ways. In the first place, models are used to estimate surfaceO_3concentrations. Furthermore, modelling activities are needed to estimate the risk posed by these O_3 levels to ecosystems or human health. Finally, modelling is essential for forecasting both O_3levels and their effects under the future scenarios of air pollution and climate change. Modelling ground-level O_3 has thus been one of the notable topics in the last decades among the air pollution community (Simpson et al., 2012).

The impact of air pollution can be indirectly estimated through epidemiological studies, the so-called "risk assessment", for both human and ecosystem health (Braun et al., 2017). Regarding human health, the risk assessment represents the number of cases of health endpoints attributable to exposure to air pollutants at any scale (World Health Organization, 2013). These results, together with information about mortality and estimated risk, can be efficiently represented on geographical maps to highlight the distribution and extent of the threat, and to identify objectives and priorities for any prevention, remediation, or mitigation actions (Briggs, 2008). Regarding ecosystem health, the generation of reliable O_3 mapsis needed for risk assessment and is a challenging process (Sicard et al., 2016b). Starting from several measurements over a large area, numerous approaches are used to map O_3 pollution and make predictions of the impacts because of O_3 pollution and have been reported in the literature (e.g.,De Marco, 2008; Li et al., 2014). Although the O_3 formation and the dispersion of its precursors in the troposphere are intricate, researchers have made significant efforts to simplify this complex behaviour and to understand the characteristics of their distribution over time.

Overall, the approachesto O_3modelling can be formally groupedinto two broad families defined as: *statistical*, which can be broken down further into "pure" statistical and geostatistical ones; and *deterministic*,e.g. chemistry transport models. Each of these two different inherently different approaches refers to variable spatial and temporal scale, with different data needs and pros and cons as highlighted in the assessment of the model performance paragraph.

The main aims of this review paper are: i) to describe the current state-of-the-art knowledge used in tropospheric O_3 modelling and mapping; ii) to discuss whether different approaches differ (or look similar) each other and identifying pros and cons for each approach; and iii) to stimulate and outline important directions for further research in O_3 modelling for the near future. Overall, the informationcollected here can be used and serving as a guideline and handbook for those approaching to study ozone and to its modellingand helping in finding the best approach for a particular purpose.

2. Statistical approach

2.1. Statistical models

Traditional statistical approaches include multiple linear regressions (Abdul-Wahab et al., 2005), multiple linear regressions combined with principal component analysis (Tan et al., 2016), and atmospheric dispersion models at city scale (Pineda Rojas et al., 2016). The multiple linear regression method was widely used because of the convenience of establishing a direct relation between O_3 and the variables associated with its behaviour, through a rather simple and explicit equation (Barrero et al., 2006). On the other hand, the non-linear relationship between O_3 and its contributing factors makes the linear models unfit (De Marco et al., 2013). Multiple linear regressions also suffer from the risk of data collinearity and limitation to obtain the best fit and therefore need large datasets of local variables (Awang et al., 2015). Furthermore, the complexity of O_3 formation, combined with the uncertainty in the measurement of most of the involved parameters, makes the modelling process intricate (Lamarque et al., 2013).

In rural areas, where the geographical distribution of monitoring stations is heterogeneous, geostatistical models are of great value (Sicard et al., 2013). Starting from a set of monitoring stations, a hybrid regression-interpolation approach was proposed, i.e., local regression followed by kriging of residuals (Sicard et al., 2016b). Land-Use Regression and ordinary kriging comprise the most used geostatistical approach for O_3 prediction (Jerrett et al., 2004) and are useful to develop optimal O_3 maps. Each geostatistical model has its inherent uncertainty due to the complexity of the atmospheric environment (Adam-Poupart et al., 2014).

2.2. Geostatistical models

The geostatistical approach assumes that the phenomena occurring in natural conditionsare spatially dependent and/or somehow correlated. Samples taken at nearby locations are expected to have more similar values than samples taken farther apart, based on the assumption that everything is related to everything else (Tobler, 1970). It was reported that Tobler's first law of geography is the core of spatial interpolation and geostatistical analysis (Miller, 2004). Spatially correlated values not only facilitate optimal and continuummapping of the pollution in the entire area but also provide valuable information about the air quality of that area (De Marco, 2008). The final objective of the spatial interpolation is to predict the air pollution concentrations over a defined region by estimating the concentrations at unmeasured locations based on known measurementsin specific sites. The simplest geostatistical methodology is the Inverse Distance Weighting (IDW), whichproduces a prediction as a weighted average of monitor data with weight based on inverse distance to the unsampled location. Berman et al. (2015) applied this methodology to map ozone concentration in the US, but its methodologieswas outperformed by other more accurate, such kriging. IDW is particularly critical because it is based on spatialautocorrelation. Indeed, IDW assigns more weight to nearby points than to distant points (Myers, 1991), thus requiring spatial autocorrelation.

Kriging is one of the first geostatistical methods used in mining and geological engineering sincethe 1950s (Chang, 2008). Since then, it has been used in air quality studies (e.g.,De Marco, 2008; Adam-Poupart et al., 2014; Sicard et al., 2016b; Feng et al., 2019), with accurate predictions and estimates (Fraczek et al., 2001). The main advantage of using kriging in spatial interpolation relies on its ability to calculate the uncertainty of prediction which is useful in decision making. A kriging interpolation model predicts surfaces better than other geostatistical models when data are checked for outliers and errors. If the data follow a normal distribution, kriging is to date the best-unbiased method of predicting a surface (Kethireddy et al., 2014), even though the spatial prediction does not necessarily require the data to be normally distributed. Technological and scientific advances led to the development of geospatial platforms in which certain tools and extensions allow studying the spatial-temporal changes of geo-environmental phenomena. It was reported that geostatistical analysis can assess potential environmental hazards by interpolating the possible flow and direction of air pollution, biohazard releases, and any potentially harmful waste that may be introduced into an area (Kethireddy et al., 2014).

An extensive analysis of different O_3interpolation techniques (from Inverse Distance Weighting to Ordinary Kriging) was performed by Hůnová et al. (2012)by estimating uncertainties linked to the interpolation asroot-mean-square error (RMSE) in Czech forests. The most suitable methodology was the ordinary kriging recommended as the optimal approach out of the eleven spatial interpolation techniques examined. The estimation of RMSE was done for both O_3concentrations and AOT40 (Accumulated hourly Ozone over Threshold of 40 ppb) and was ranging between 10% and 20%, respectively (Hůnová et al., 2012). Another example of geostatistical methodologies used to map $O_3$was applied by Frazcek et al. (2001) in the Sierra Nevada (United States) and Carpathian (Central Europe) Mountains. In particular, kriging and co-kriging were compared. The latter was performed using two additional variables related to O_3concentrations, elevation, and maximum air temperature. The use of the additionalvariables was able to supply the low intensity of O_3 data in the CarpathianMountains. Sufficient numbers of monitoring sites, spatially homogeneous distributed across the territory, werefound to be a key factor for model accuracy and reliability (Frazcek et al., 2001; Sicard et al., 2016b).

Amongthe different kriging options, Universal kriging, was observed to give better results than Ordinary krigingin US ozone concentrations mapping, allowing to assess the significance of environmental covariates for both inference and prediction of O_3concentrations (Berman et al., 2015). Between the concerns of kriging, it is depending on spatial autocorrelationas IDW. Indeed, a critical component of kriging is the semivariogram. A useful semivariogram cannot be developed without the presence of spatial autocorrelation; the degree of spatial autocorrelation determines how successful spatial interpolation will be (Griffith and Layne, 1999).

Kriging performance can be improved using co-variable related to ozone concentrations in the co-kriging interpolation. Co-kriging allows to better estimate primary variable if the distribution of a secondary correlated variable is sampled more intensely than the primary variable. The most used co-variable is the Digital Elevation Model (DEM), taking into account topographic effects (Sicard et al., 2013). The authors applied this methodology in Mediterranean basin to background ozone stations. The co-kriging was able to compensate for the lack of sufficient sampling in some areas. The RMSSE was always close to 1, with 1 highlighting no variability in prediction and thus no uncertainty for the whole domain.

Land-Use Regression models.

More advanced exposure estimation techniques include Land-Use Regression (LUR) models. For instance, LUR models are used to predict air pollutant concentrations at unmonitored sites based on regression models of geo-referenced covariates that predict observed data in monitoring sites (Beelen et al., 2009). LUR modelling employs statistical methods to combine data from air pollution measurements with data from Geographic Information Systems to explain spatial concentration variations (Hoek et al., 2008). A LUR model can characterize the spatial variability of air pollutants considering other information, such as roadside dispersion profiles. The model performance is limited by the number and the spatial distribution of sampling sites (Basagana et al., 2012; Wang et al., 2012). Kerckhoffs et al. (2015) applied LUR to predict O_3 levels in the Netherlands. They found that O_3 levels were highly correlated with NO and moderately with fine particles. They built a LUR model including small-scale traffic, large-scale address density, urban green and a regional indicator that was able to explain 71% of the spatial variation in summer average O_3concentrations.

2.3. Machine Learning algorithms

Spatial linear LUR is commonly used for long-term modelling of air pollution in support of exposure assessment. However, Machine Learning methods, with spatio-temporal modelling, provide more accurate exposure metrics than LUR in modelling human exposures for epidemiological studies (Ren et al., 2020). Ren et al. (2020) and Requia et al. (2020) have compared different Machine Learning algorithms to model the relationship between dependent variables and predictor variables to fill in the missing values, in order to estimate the daily maximum 8-h mean O_3 concentrations at high spatial resolution (1×1 km grid cell) across the United States: linear regression models (i.e., Multiple Linear Regression, Ridge regression, Elastic Net regularization, Principal Component Regression, Partial Least Squares Regression) and non-linear modelling options (i.e., Lazy Learning, k-Nearest Neighbors, Kernel Trick, Support Vector Regression, Artificial Neural Networks, Artificial Neural Networks, Deep Neural Networks, Decision Trees, Regression Trees, Random Forest, and Extreme Gradient Boosting).

The non-linear Machine Learning methods led to higher accuracy of predictions compared to linear LUR i.e., 10–40% decrease of predicted RMSE (Ren et al., 2020). By applying three Machine Learning algorithms (neural network, random forest, and gradient boosting) Requia et al. (2020) obtained high model performance ($r^2 = 0.86$–0.90), and the best performance was observed during summer ($r^2 = 0.88$) in the United States. The performance of the Machine Learning algorithms depends on the location and O_3concentration, therefore it recommended to apply a hybrid model instead of a single model (Ren et al., 2020).

2.4. Deterministic approach

Chemical transport models.

Because O_3 is a highly reactive trace gas, estimates of its contribution to climate forcing must rely on global chemistry-transport models (Derwent, 2020). Atmospheric chemical transport models (CTMs) are used to simulate the formation, removal and transport of O_3 (Lamarque et al., 2013) into the troposphere. These models were formulated to quantify the impact of air pollutant emissions on the chemical composition of the atmosphere and corresponding consequences on the environment (Gupta and Mohan, 2015). The reliability of the models increases with increasing temporal and spatial resolution of input data, such as emission inventory (Karlický et al., 2017). The key limitations of CTMs include the requirements of high computational resources and data, and good knowledge about the atmospheric processes and source of air pollution (Tong et al., 2011). Furthermore, CTMs tend to underestimate the magnitude of fluctuations on shorter temporary scales with the possibility of overestimating during periods of extensive cloud cover (Pal et al., 2014). Many studies have used computer numerical models implemented at regional (or even global) scales to supplement the missing information from in-situ measurements (Sicard et al., 2021b). In the past decades, the importance of these numerical models has been increasingly recognized and numerous air dispersion or air quality models were developed at various spatial scales to assist in understanding, controlling, and forecastingair pollution (Miranda et al., 2015). CTMs are numerical models that simulate over a given region the atmospheric chemistry taking into account four main processes: i) assessment of natural and anthropogenic emissions ii) atmospheric transport, iii) chemical production/destruction and iv) losses to surface by dry deposition. They are widely used to estimate the concentration of gases in the atmosphere at different temporal and spatial scales and have been successfully applied to air pollution research and air quality management at a regional scale worldwide (e.g.,Reis et al., 2005; Haase et al., 2014; De Marco et al., 2020).

CTMs have been used in China, covering wholecountry or specific regions (Hu et al., 2015; Li et al., 2018; Quennehen, 2008). However, the representation of air quality and meteorology over Asia is still challengingbecause of complex physical and meteorological conditions of this area, characterized by the monsoon system and large uncertainty inthe anthropogenic emissions (Sicard et al., 2021b). Nevertheless, CTMs are widely used to study air quality over Asia. For instance, Hu et al. (2015) applied the Community Multi-scale Air Quality (CMAQ) and Weather Research Forecasting (WRF) modelling system to predict air pollutant concentrations for the whole of China. The results showed an overestimation of 1-h or 8-h O_3 average, due probably to the coarse horizontal resolution (36 km). A modified WRF/CMAQ experiment was performedto simulate O_3 in winter (December 2014–February 2015) and summer (June–August 2015) for the entire Sichuan Basin (Qiao et al., 2019). The 1-h and 8-h O_3 averages were both greatly over-predicted in winter, but the model performance was acceptable in summer when the photochemical production of O_3since anthropogenic emissions should be strongest in the basin (Qiao et al., 2019). The WRF-CMAQ model was used in India with a different spatial resolution for emissions and meteorological inputs (e.g., 36 km) to assess source and species sensitivities of ground-level O_3 concentrations (Sharma et al., 2016). In the framework of the Hemispheric Transport of Air Pollution (HTAP) phase 2 experiment, simulations of O_3 and its precursors were conducted using the updated version-2 (HTAP-v2) emission inventory and the offline global chemistry transport model MOZART-4 (Surendran et al., 2015). Comparison between model simulations and surface/balloon-borne observations at several sites showed reasonable model performance, but some disagreement in O_3 concentration and seasonal variation over South Asia was still evident (Surendran et al., 2015). The WRF model, coupled with Chemistry (i.e., WRF-Chem) was used to simulate the spatial and seasonal variability of main physical and chemical variables over Asia for the year 2015 at 8-km horizontal resolution and to estimate O_3impacts on Asian forests (De Marco et al., 2020). Overall, WRF-Chem reproduced well the spatial and seasonal variability of tropospheric O_3 content, with limited overestimation during the warm season (3–7%) and larger over-prediction (11–13%) during the cold period (Sicard et al., 2021b). Emissions ofNOx, methane, carbon monoxide and isoprene had the potential to contribute in a major way to model output uncertainties (Derwent, 2020).

In Europe, CTMs are used with resolution ranging between 12 and 25 km for operational European wide applications (e.g.; Mues et al., 2014; Anav et al., 2016), 4–10 km for application to a single country (e.g. Vieno et al., 2010; Baldasano et al., 2011; Hendriks et al., 2013) and reaching 1 km for some European regions (Pay et al., 2014). As an increase in horizontal model resolution will quadratic or cubically increase the computational costs and poses additional challenges concerning high-resolution input data and model formulation, it is important to reach a cost-effective compromise (an "optimum resolution") in the trade-off between the model performance and computational cost. Projected changes in ground-level O_3 vary considerably among models (Wild et al., 2012). Several studies have evaluated the impact of spatial model resolution on O_3 production (e.g., Tie et al., 2010; Lauwaet et al., 2013) as well as O_3 precursors (Wałaszek et al., 2018). In general, high-resolution simulations may provide a much better separation between regions defined by high concentrations of biogenic volatile organic compounds and high NOx levels (Pugh et al., 2013). Tie et al. (2010) performed an experiment over United States to assess the impact of model resolution on simulated air quality; they used a domain of 36, 12, and 4 km^2respectively, finding that the 36-km^2 resolution leads to an under-prediction of daily maximum 8-h O_3 averages, and an over-prediction of daily minimum 8-h O_3 averages (Tie et al., 2010). Otherstudies support the finding that modelled O_3 formation systematically increases with the resolution for regional and global scale applications (Wild and Prather, 2006). Evaluations of global, hemispheric, and regional CTMs show that regional models typically perform better (van Loon and Coauthors, 2007; Simpson et al., 2014).

In Europe, long-term O_3 simulations from seven regional air quality models (i.e., the Unified EMEP model, LOTOS-EUROS, CHIMERE, RCG, MATCH, DEHM, and TM5)were inter-compared and compared to O_3 measurements within the framework of the EuroDelta experiment (van Loon and Coauthors, 2007; Colette et al., 2011). This study clearly showed that increasing model resolution is advantageous for European scale applications and that moving from a resolution of 50 km in favour of a resolution between 10 and 20 km is practical and worthwhile. With increasing resolution of the meteorological model and emission inventories, andadjustment of CTM process descriptions and parameterizations to this higher resolution, an improved performance of CTMs model is expected (Schaap et al., 2015). The performance of the different models to simulate O_3 fields is compared and in general the models reproduce the main features of the O_3 diurnal cycle, even if overestimating daytime O_3. LOTOS-EUROS and RCG have a more pronounced diurnal cycle variation than observations, in contrast with TM5. CHIMERE has a large positive bias, which can be explained by a systematic bias in boundary conditions.

Regional chemistry-climate models at coarse horizontal resolution (e.g., 36–50 km) are often unable to resolve the local features influencing the chemical transformation and poorly reproduce the ground observations (Schaap et al., 2015). On the other hand, a weakness linked to regional models is their high time and resources consumption. Indeed, the run of a single model requires from weeks to months to have a final output.

In recent years, researchers have focused their attention on advanced models like ensemble models, which showed better performance than standard single CTMs (Gong &Ordieres-Meré, 2016). Singh et al. (2013) used the ensemble trees to predict air quality, applying meteorological parameters as estimators. The methodology to assess the model performance was based on classification and regression.

Recently a new approach was developed to integrate CTM predictions and measures, the so-called Regionalized Air Quality Model Performance approach, using the Bayesian Maximum Entropy framework (DeLang et al., 2021). Thus, estimates are produced that put priority on observations and take advantage of air quality model predictions based on how well they reproduce the observed values. Spatial fields generated from this approach provide an observation and CTM informed representation of O_3 across space/time that is more accurate and precise than relying only on observation data. This was especially true for locations away from monitoring stations.

2.5. Chemistry-climate models

In the climate models, physical atmospheric processes are calculatedby solving equations that describe fluid flow and radiative transfer, which can not only respond to changes in greenhouse gas concentrations, solar output, or other forcing but also generate their internal meteorological variability (Flato et al., 2014). Chemistry-climate models, or composition-climate models (CCMs), represent the most complex models in this family, where the chemically driven changes in radiatively active gases and aerosols (e.g. O_3, methane, sulfates) influence the model radiation scheme, thus chemistry directly influences climate through its direct and indirect effects (O'Connor et al., 2015). The use of CCMs with tropospheric chemistry and aerosols is a relatively recent development (Morgenstern et al., 2017), whereas coupling of upper atmosphere chemistry to climate has a much longer history due to the increased importance of chemically active compounds for heating rates in the stratosphere (Morgenstern et al., 2017). A less complex model than the CCM is the chemistry CTM (Chemical Transport Model), where the chemistry is affected by the climate changes from the radiative and dynamical parts of the model but the chemically driven changes in radiatively active gases and aerosols do not subsequently affect climate. This type of model was the first step in coupling tropospheric chemistry to physical climate models (Roelofs and Lelieveld, 1997), and it is still occasionally used (Lamarque et al., 2013).

2.6. Chemical reanalyses and ensemble approaches

Applications of chemical ensemblesinclude comprehensive spatio-temporal evaluation of independent models, such as those developed in the framework of the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP; Yang et al., 2012) and CCMI (Morgenstern et al., 2018). In their study the ACCMIP ensemble O_3 simulations were evaluated using a chemical reanalysis, complementing the use of individual measurements for such a purpose. The chemical reanalyses can also be used as an input to meteorological reanalyses, as for radiation calculations (Dee et al., 2011), and can provide boundary conditions to regional-scale models and to analyze particular pollution events such as those associated with heatwaves or large-scale forest fires (Huijnen et al., 2012). Finally, they can be used as a reference to identify to what extent particular periods and regions deviate from climatology, as provided by the reanalysis, for instance as also discussed in the series of the “State of the Climate” (Flemming and InnesA, 2018).

The global chemistry models developed under the ACCMIP project were used by Sicard et al. (2017) to determine the impacts of O_3on forests productivity at the global level, in four climate scenarios RCPs. ACCMIP models were widely validated and used to evaluate projected changes in atmospheric chemistry and air quality under different emission and climate assumptions (Lamarque et al., 2010; Voulgarakis et al., 2013). Lamarque et al. (2013) provided the main characteristics of 16 models and details for the ACCMIP simulations. The length of historical and RCP simulations varies between models, but for all models, the historical runs cover a period centred around 2000, while the time slice of RCPs is centred around 2050 and 2100.

Changes in emissions from one region can impact air quality over others, also affecting air-pollution-related health impacts due to intercontinental transport (Zhang et al., 2017). In the framework of the Task Force on Hemispheric Transport of Air Pollution (TF-HTAP), Anenberg et al. (2009) found that reduction of foreign O_3 precursor emissions can contribute to more than 50% of the avoided deaths by simultaneously reducing both domestic and foreign precursor emissions. A reduction in emissions in North America and Europe results in largest impacts as reduction of O_3-related premature deaths in downwind regions as compared to within the source region (Anenberg et al., 2009).

For most O_3 indicators, the ensemble average identified “ensemble model”, almost always exhibits a superior skill compared to any individual model, even though it has a too weak variability. The spread of ensemble-model values is fairly representative of the uncertainty of summertime O_3 daily maxima, as the occurrence of the observation within the model values range has a rather flat distribution, when the bias is removed. For a given day, the probability distribution of occurrence of the observation is well represented by the distribution of the simulated values.

2.7. Assessing model performance

Statistical models are associated with a relative simplicity of the approaches suggested, even though they are associated with high level of uncertainties (LRM) and high RMSE (IDW, Kriging). Methodology with increasing level of complexity, such as co-Kriging, LUR and Machine Learning, showed lower RMSE, but on the other hand have much more information as input data requested. All the statistical models requires measured data, and have as output continuous layers of pollutant concentration over a specific domain. This request imply that the RMSE is lower when the distance between measurement sites is lower, decreasing in this way the uncertainties of the modelling approaches.

The deterministic models are generally characterized by lower level of accuracy and higher accuracy, showing different level of complexity, that is increasing from Global model to Regional models. These type of models require high number of input data, such as emission inventories, and need pollutant dispersion module and meteorological model. On the same time they request high computation time and high storage room.

Validation of the model performance from ground observations is, so far, still a problem due to the scarcity of monitored information (Sicard et al., 2021b). Recently, few authors have validated regional model data with in-situ, balloon-borne observations, and satellite observations (e.g., Im et al., 2015; Surendran et al., 2015; Ghim et al., 2017; Crippa et al., 2019; Sicard et al., 2021b). The model performance can be evaluated over different seasons by using the Pearson’s correlation coefficient (r), mean bias (MB), the fractional bias (FB), and the Root-Mean-Square Error (RMSE). These statistics are successfully used in several studies for evaluating the performance of regional air quality models (e.g., Im et al., 2015; Ghim et al., 2017; Crippa et al., 2019; Sicard et al., 2021b). In case of in-situ data, we have to extract model results at the lowest model layer, and we calculate the performance statistics for each station. The Pearson’s coefficient allows estimating the spatial agreement between model and observations. For physical parameters, the MB provides the absolute bias of the model, with negative and positive values indicating respectively underestimation and overestimation by the model while the FB (in %) is used for the chemical variables, as in this case the absolute bias would be hard to interpret. The cross validation (e.g., 10-fold cross validation) is usually used. Here, we first divide the monitoring sites into 10 splits, and then we train the model with 90% of the data and predicted the O_3 concentration at the remaining 10% of the sites. Then, the RMSE is commonly used to measure the differences between modelled values and the observations.

We have summarized all pros and cons of the different approaches in Fig. 1, where in the same time we estimated the performance of each approach on the base of its accuracy, spatial resolution, complexity, temporal resolution, statistical vs. deterministic nature and data need. We gave a score to each of these parameters ranging between 0 and 1 and at the end we have estimated the performance of the model on the basis of the area of the obtained graphs. Higher is the area, higher is the performance of the model.

3. Ozone risk assessment

3.1. Models for human health

Acute exposures are characterized by high O_3 concentrations for a relatively short-time period, within hours or days, while chronic exposures involve lower O_3 concentrations persisting, or recurring, over a longer period (Grulke et al., 2007; World Health Organization, 2008; Sicard et al., 2016b). To protect population, it was considered that the 8-h guideline would protect against acute elevated 1-h O_3 exposures (World Health Organization, 2008). The current O_3human health metrics (SOMO35, i.e., the annual Sum Of daily maximum 8-h Means Over 35 ppb, and the number of exceedances of daily maximum 8-h values greater than 60 ppb) consider only acute health effects (e.g., lung inflammation), and do not account for possible chronic effects at long-term O_3 exposure levels below 35 ppb (World Health Organization, 2013).

In cities, $PM_{2.5}$ and ground-level O_3 have potentially the most significant adverse effects on human health associated with respiratory and cardiovascular diseases and mortality, compared to other air pollutants (World Health Organization, 2013; Cohen et al., 2017). The Global Burden of Disease (GBD) Study reported 4.1 million disability-adjusted

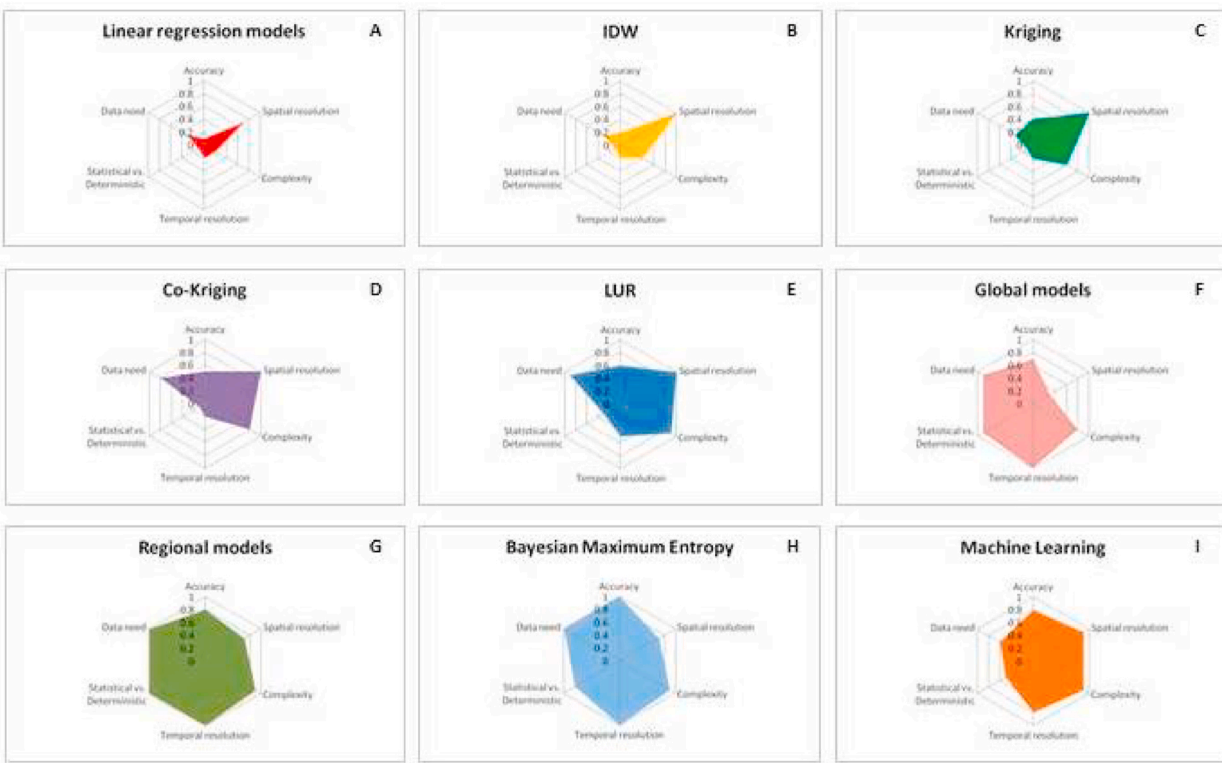

Fig. 1. Performance of the different approaches. A score was assigned on the basis of the characteristics of the suggested approach in terms of accuracy, spatial resolution, complexity, temporal resolution, statistical vs. deterministic nature and data need. The higher is the area the higher is supposed to be the model performance and its applicability.

life years (DALYs) in 2015 attributable to O_3 exposure (Forouzanfar et al., 2016) that is estimated to provoke more than 0.7 million deathsper year worldwide (Anenberg et al., 2010). Short-term effects induced by the oxidative stress of this pollutant on the respiratory system are well-established, particularly in people with pre-existing obstructive chronic pulmonary diseases (COPD) (Nuvolone et al., 2018). The World health organization (World Health Organization, 2013), theEuropean Council (Directive, 2008/50/EC), the Environmental Protection Agency (US Federal Register, 2015) have set Ambient Air Quality Standards for the protection of human health (Table 2). For ground-level O_3, China adopted in 2012 the Ambient Air Quality Standard of 80 ppb as maximum daily 8-h running average (Ministry of Environmental Protection, 2012) (Table 1).

For the estimation of health effect, different methods or models are used. The more common methods for assessing the short-term effects of air pollution on human health is time-series analysis. The time-series analysis method is a simple and descriptive-statistical approach for O_3 modelling (Tian et al., 2020; Javanmardi et al., 2018). Time-series analyses showed that O_3 is associated with an increased risk of premature mortality, but currently, statistical models (including meta-analysis regression) are being developed to establish indicators of risk mortality or hospital admission from short-term O_3 exposure (e.g. maximum daily 8-h mean concentration). Li et al. (2020) used two stages strategy to investigate the relationship between O_3 exposure and years of life lost (YLL). For this, city-specific associations were calculated by generalized

Table 1
Air quality standards for ozone in terms of target values for the protection of human health (World Health Organization, 2013; Directive, 2008/50/EC; US Federal Register, 2015; Ministry of Environmental Protection, 2012).

Guideline	WHO	EU	US	China
Maximum daily 8-h mean (ppb)	50 ppb	60 ppb	75 ppb	80 ppb

Table 2
Relative Risk for health outcomes, and people at risk (e.g., all ages or > 30 years–old), for 10 μg m^{-3} increase in daily maximum 8-h ozone mean-concentrations with 95% confidence intervals (lag 0–1 days).

Outcome - Region	RR (95% CI) per 10 μg/m3	Ref.
Mortality		
All-causes (all ages) - Europe	1.003 (1.001–1.004)	(WHO, 2013)
All-natural causes (>30 years) - France	1.009 (1.004–1.014)	Sicard et al. (2019)
All-cause mortality, summer (all ages)– United States	1.007 (1.004–1.011)	Bell et al. (2005)
Cardiovascular diseases (all ages) - Europe	1.005 (1.002–1.007)	Gryparis et al. (2004)
Cardiovascular diseases (>30 years)- Europe	1.004 (1.003–1.005)	(WHO, 2013)
Respiratory diseases (all ages) - Europe	1.013 (1.007–1.015)	Gryparis et al. (2004)
Respiratory diseases (>30 years)- Europe	1.014 (1.005–1.024)	Héroux et al. (2015)
Daily Hospital Admissions		
Chronic Obstructive Pulmonary Disease (all ages) - Europe	1.009 (1.004–1.013)	(WHO, 2013)
Cardiovascular diseases (all ages) - Europe	1.009 (1.005–1.013)	Héroux et al. (2015)
Respiratory diseases (all ages)- Europe	1.004 (1.001–1.008)	Héroux et al. (2015)
Respiratory diseases (15–64 years old) - Europe	1.001 (0.991–1.012)	(WHO, 2008)
Respiratory diseases (≥65 years old) - Europe	1.005 (0.998–1.012)	(WHO, 2008)
Myocardial infarction (ozone <70 ppb)– United States	0.998 (0.996–1.000)	Yazdi et al. (2019)
Pneumonia (ozone <70 ppb) – United States	1.030 (1.028–1.032)	Yazdi et al. (2019)

additive models (GAM). The family function of GAM is the Gaussian model (Guo et al., 2013). The daily observed YLL (95% CI) was estimated by the following equation (1), and their sum divided by total non-accidental mortality was additional life gained per deceased people.

$$\sum_{t=1}^{1826} (YLL_t) \times ([O_3]_t - Target) \times \beta \quad (1)$$

where YLL_t and $[O_3]_t$ are respectively the daily number of lost life years and the O_3 concentration at the day t. Target and β are the concentration target of ozone and the national average relative change of YLL per 1 μgm^{-3} increase of ozone, respectively (Li et al., 2020).

These statistical models relate mortality or hospital admission to data on air pollution, and weather, correcting by age, gender, socio-economic indicators, race/ethnicity, etc. (Zanobetti and Schwartz, 2008). For estimation of mortality and hospital admissions due to O_3 exposure some models need inputs such as baseline incidence (BI), relative risk (RR), specified population and 1-h or 8-h concentrations of O_3. The BI use epidemiological models to estimate the occurrence of events in different population subsets like the evidence of events for patients with risk factors compared with those without risk factors (Nelson et al., 2015). Relative risk is the possibility of developing a disease following exposure to a pollutant. The RR is the attributable health risk related to people who have defined exposures and so it can be calculated by the equation (2).

$$RR = \frac{\text{Probability of a health effect when exposed to air pollution}}{\text{Probability of a health effect when not exposed}} \quad (2)$$

The estimation of different diseases and mortality due to O_3exposure is based on BI and RR, calculated from meta-analysis and/or epidemiologcal studies for hospital admissions and mortality due to cardiovascular and respiratory diseases (World Health Organization, 2013). Table 2 shows the different RR in the studies of ozone exposure and its effects on human health.

Several models have been used to investigatethe effects of O_3 exposure on human health (Sicard et al., 2019; Yang et all, 2012; Jerrett et al., 2009; Gryparis et al., 2004). For instance, the AirQ software elaborated by the WHO Regional Office for Europe is widely used worldwide e.g., in Asia (Yang et al., 2020), Iran (Khaniabadi et al., 2018; Amoatey et al., 2019), and Europe (De Marco et al., 2009; Sicard et al., 2019; Khaniabadi and Sicard, 2021). In this AirQ model the BI and RR are used as input for estimation of mortality and hospital admissions. The attributable proportion AP, defined as the fraction of health consequences in a population exposed to ozone, is calculated as follows:

$$\mathrm{AP} = \sum\{[\mathrm{RR(c)} - 1] \times \mathrm{P(c)}\} \Big/ \sum[\mathrm{RR(c)} \times \mathrm{P(c)}] \quad (3)$$

Where RR(c) is the relative risk of a certain effect on health in category "c" (e.g., residential or industrial) of exposure that it is taken from the exposure-response functions from published epidemiological studies, being P(c) the number of individuals in the under exposure population. Table 3 shows some results of studies, published in the last decade, using AirQ model for estimating mortality due to O_3 exposure in different countries. Almost all papers investigated cardiovascular and respiratory-related diseases with the RR suggested in Table 2.

The O_3 concentrations increased in most cities worldwide (on average: + 0.31 ppb $year^{-1}$ since 1990s), in particular + 0.33 ppb $year^{-1}$ in North America, + 0.68 ppb $year^{-1}$ in East Asia and +0.27 ppb $year^{-1}$ in Europe between 2005 and 2014 (Sicard, 2021). Between 2000 and 2017, the annual O_3-related number of premature deaths increased (+0.55 deaths per 10^6 inhabitants) in the EU-28 cities (Sicard et al., 2021a). Knowlton et al. (2004) assessed changes in O_3-related mortality in the 2050s compared with the 1990s using the coupled GCM/MM5/CMAQ models in the United States, and in the New York state. They found that climate change could increase regional summer O_3-related mortality by a median of 4.5% in the 2050s compared with the 1990s, without including the populationgrowth, reaching a median of 59.9% when the increase in population at risk was considered.

4. Models for plant ecosystems

To protect vegetation, the current exposure index AOT40 assumes O_3 concentrations below 40 ppb, and do not account for possible chronic effects (Sicard et al., 2016b).

To provide accurate impact metrics to protect human health and vegetation from adverse effects of O_3, accurate hourly O_3 concentrations are needed, and the models must reproduce well the spatial and seasonal O_3 variability even for lower concentrations e.g., in cities. The performance statistics have to be based on hourly data year-round.

Apart from complementing atmospheric chemistry measurements and evaluating potential threats to human health, modelling activities are also applying to estimate the risk posed by these O_3 levels to ecosystems, particularly to vegetation. The adverse effects of O_3 on plants were first identified in the 1950s (Ashmore, 2005). Ozone can reduce agricultural yield, mainly by the appearance of visible injury that reduces market value and the reduction of the yields due to decreased photosynthetic rates and accelerating leaf senescence (Feng et al., 2019; González-Fernández et al., 2016; Monga et al., 2015; Pleijel et al., 2019). There is also evidence of lower growth of tree species, reductions of the nutritional quality of pastures, reduction in the reproductive capacity of plants, and biodiversity changes in the species composition of pastures (Agathokleous et al., 2020; Alonso et al., 2014; Ashmore, 2005; Büker et al., 2015; Li et al., 2018; Sanz et al., 2016).

The methodology for vegetation risk assessment was developed in the framework of the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP). Experimental studies in which vegetation is exposed to

Table 3
Number of cases attributed to exposure to ozone (O_3) for mortality for all natural causes, cardiovascular diseases, and respiratory diseases, using AirQ software (95% Confidence Interval).

Reference	Location	Time period	Annual mean conc. (μg/m^3)	Ozone-related premature deaths		
				All-causes	Cardiovascular diseases	Respiratory diseases
Rovira et al. (2020)	Catalonia, Spain	2017	59.8	–	–	9 (3–15)
Bonyadi et al. (2020)	Shirza, Iran	2017	51.6	122 (8–202)	85 (34–119)	45 (25–72)
Du et al. (2019)	Jinhua, China	2019	84.5	–	–	–
Amoatey et al. (2019)	Ahvaz, Iran	2015	79.2	128 (85–171)	156 (0–233)	41 (21–61)
Sicard et al. (2019)	Marseille, France	2015	55.0	169 (76,261)	65 (12–117)	20 (10–32)
Asl et al. (2018)	Hamadan, Iran	2014–2015	73.0	52 (35–86)	36.5 (15–50)	19.1 (11–22)
Sicard et al. (2019)	Rome, Italy	2015	41.5	614 (372–853)	69 (51–86)	37 (14–59)
Khaniabadi et al. (2018)	Kermanshah, Iran	2014–2015	83.2	–	83 (0–123)	32 (13–53)
Hadei et al. (2013)	Tehran, Iran	2015–2016	42.1	341 (228–565)	272 (122–419)	123 (68–142)
Jeong (2013)	Suwon, Korea	2011	43.0	43 (29–71)	16 (6–22)	14 (7–16)

different concentrations of ozone are used to obtain a relationship between concentration of pollutants in the air and their effects on plants (CLRTAP, 2017). When just the concentration (exposure) is used to establish this relationship, the atmospheric chemistry simulations alone are sufficient to assess the risk that O_3 poses to vegetation at regional scale, testing the modelled concentrations against established O_3-level thresholds. This is a very useful approach for large scales since it is possible to apply for most vegetation types (Sicard et al., 2019). However, in the framework of the CLRTAP, vegetation risk assessment is preferably performed by calculating the exceeded dose of O_3 absorbed by the plant stomata over anexperimentallyset threshold, the so-called phytotoxic ozone dose (POD) (CLRTAP, 2017). For POD, two steps are required: a CTM accounting for the surface concentrations and a parameterization which allow to estimate the vegetation absorption of O_3 (occurringthrough plant stomata).

Resistance-analogue models are commonly applied to estimate ozone transfer from the atmosphere to vegetation. One of the most used models to estimate ozone uptake by plants is the DO_3SE model (Deposition of O_3 for Stomatal Exchange; Jarvis, 1976; Büker et al., 2012), a multiplicative algorithm. DO_3SE is currently included within the European Monitoring and Evaluation Programme (EMEP; Simpson et al., 2012) photo-oxidant CTM, which simulations are used in the framework of CLRTAP to inform the European strategies for pollutant emission control (Simpson et al., 2007). However, for using this method, the threshold for different plant species or vegetation types must be defined, as well as the species-specific parameterization of the DO_3SE model. De Marco et al. (2020) applied a CTM over Asia, to assess the O_3 risk for forests, comparing both metrics to estimate potential ozone damage: exposure- and flux-based. Using POD allowed including in this analysis some limiting conditions of plant activity, such as soil water content, which is key for risk assessment in a context of climate change. In Europe, the application of the DO_3SEmodel to calculate POD in thecontext of climate change allowed toalso consider the implications of a changing ozone concentration profile in future scenarios (Hayes et al., 2019).

The above-exposed methodologiesare empirical approaches to modelO_3effects that can be completed with other empirical plant-response functions and applied to different environmental conditions (De Vries et al., 2017). However, new modelling approaches based on mathematical simulations of the processes behind the pollutant effects are currently being developed (Kinose et al., 2020; Schauberger et al., 2019). These process-based models are considered veryreliable tools for simulating the effects of O_3on biological dynamics on novel conditions including air pollution and climate scenarios (Evans, 2012) that can complement the current approach.

5. Conclusive remarksand future directions

Due to the impossibility to measure surface O_3concentrationsin large regions, for economic and practical reasons, the use of modelling is very important and highly suggested methodology to define the risk assessment for ecosystems and human health. In general, the geostatistical approach is relatively simple, requiring low informatics and storage resources. It is strongly limited by the availability of data on the study area, but this limit can be minimized by the use of the co-kriging methodology, where other variables, spatially and temporally correlated with O3 concentration, can be used. Many different types of models are available to map appropriately tropospheric O_3starting from the simple geostatistical model to more complex CTM. The choice of a specific model instead of another one dependsonthe available data and onthe final objective of the investigation. Indeed,O_3 risk assessment can be obtained at global, regional or local scale using different resolution levels. Increasing the resolution level can increase the uncertainties level, but a moderate resolution improvement can improve the information released. While geostatistical model use is not the time or computation resources needed, they can be considered as "quick", but the ozone distribution in areas not covered by measurements is highly uncertain with ordinary methodology and less uncertain when co-variables are used.

Author contributions

ADM designed the structure, **MV** and **CP** authored first draft of the geostatistical approach, **HGG** authored the first draft related to plant risk assessment, **YOK** and **PS** authored the part related to human health risk assessment, **ADM**, **AC**, **ZZ**, **PS** and **EP** critically revised the manuscript. All authors revised the manuscript for important intellectual content and approved the final version for submission.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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