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3 **Chemical Weather and Chemical Climate**

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5 Guy P. Brasseur^{1,2}

6
7 Rajesh Kumar¹

8
9 ¹National Center for Atmospheric Research, Boulder, CO.

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11 ²Max Planck Institute for Meteorology, Hamburg, Germany

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15 **Abstract**

16 Progress in fundamental understanding of atmospheric chemistry in the past decades has enabled
17 the development of advanced environmental predictive capability. Accurate chemical weather
18 forecasts and source attribution information can play a decisive role in mitigating the exposure of
19 the public to acute air pollution episodes in the short-term. Chemical climate projections will
20 allow investigation of the long-term consequences of human choices and their impact on the
21 future evolution of the Earth system. This commentary stresses the importance of integrating
22 atmospheric chemistry in operational environmental prediction and projection systems.

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24 **Plain Language Summary**

25 Advances in fundamental knowledge and the availability of frontier technologies to observe and
26 simulate the atmosphere have enabled the development of comprehensive *chemical weather*
27 forecast systems at the global, regional, and local scales. Such prediction capability can play a
28 decisive role in mitigating the exposure of the public to air pollution and in identifying the major
29 sources of air pollutants. Longer-term *chemical climate* projections that integrate
30 interdisciplinary information from atmospheric chemistry, biogeochemistry, meteorology,
31 oceanography, ecology, economics, and social sciences allows the investigation of the long-term
32 consequences of human choices and their impact on the future evolution of the Earth system.
33 Time has come to move beyond purely exploratory approaches and establish prominent
34 operational projects, specifically in low- and medium-income countries (1) to improve local air
35 pollution forecasts based on methodologies that bear resemblance with those adopted in
36 meteorology and (2) to develop integrated environmental and climate prediction systems that

will assess the response to human activities and the potential success of proposed mitigation strategies.

Key points

Atmospheric chemistry should be included in operational environmental prediction and projection systems.

Chemical weather forecasts and source attribution information play a decisive role in mitigating the exposure of the public to air pollution.

Chemical climate projections that integrate interdisciplinary information allows the investigation of the consequences of human choices.

1. Introduction

Meteorological Weather refers to the state of the atmosphere and its day-to-day variability, while *climate* refers to the “average weather” over a period of typically 30 years, although, depending on the application, this period may range from months to millennia and beyond. Both cases examine physical quantities including temperature, pressure, wind, moisture, cloudiness, and precipitation, although climate broadly represents the state and long-term evolution of the coupled atmosphere-ocean-land system. Recently, the definition of weather and climate have been broadened to include disciplines other than meteorology. For example, space physicists introduced the term *space weather* in the 1950s (Gold, 1959; Kane, 2006; Cade and Chan-Park, 2015) to describe rapid fluctuations in solar radiative and particle emissions and their effects on the Earth's magnetosphere, ionosphere, and thermosphere.

About 20 years ago, the atmospheric chemistry community introduced the term *chemical weather* to describe the short-term (minutes to days) variations in the atmospheric chemical

composition (trace gases and aerosols). Lawrence *et al.* (2005) indicates that this notion “has arisen from the recognition of the tremendous short-term variability of the atmospheric chemical composition, which results from the strong influence of meteorological variability, chemical complexity, and regionally and temporarily varying emissions”. Rapidly changing emissions from erratic wildfires, temperature and humidity dependent biospheric processes, and economic activities contribute substantially to variability in the atmospheric composition. *Chemical climate* refers to long-term (decades to millennia) evolution of the atmospheric chemical state and is controlled by changes in anthropogenic emissions, physical climate, and ecosystems.

Atmospheric chemistry not only describes the atmospheric chemical composition, but also identifies the sources and sinks of atmospheric chemicals, quantifies their chemical transformation and assesses their response to natural and human forcing. This field has evolved rapidly since mid- nineteenth century through different milestones: (1) the discoveries of the atmospheric chemical elements; (2) the mechanisms producing secondary atmospheric chemicals; (3) the development of advanced multi-platform instrumentation including satellites to monitor atmospheric chemicals; (4) the development of increasingly complex numerical atmospheric chemistry models; (5) the assessment of long-term human impacts on atmospheric parameters including acidic precipitation, ozone depletion, and the atmospheric oxidizing power; (6) the development of chemical weather and chemical climate prediction capabilities based on understanding gained through systematic field campaigns; and (7) the connection between atmospheric chemistry, human health, food security, and water cycle. While some fundamental aspects of atmospheric chemistry still remain unknown, time has come for the discipline to move beyond purely exploratory approaches and establish prominent operational projects focusing on air quality prediction and analysis in ways that bear resemblance with the approach used in meteorology.

2. The Exploratory Period: Investigation of Chemical Processes

Similar to meteorology, observations and measurements provided the first quantitative advances in atmospheric chemistry. In the nineteenth century, the fathers of modern chemistry developed innovative chemical measurement methods, which led to the discovery of new substances

including ozone (Brasseur, 2020). The peculiar smell perceived in 1839 by Schönbein (1840) during the electrolysis of acidulated water was attributed only 25 years later by Soret (1865) to the existence of a molecule composed of three oxygen atoms (OOO). This gas, identified as a permanent atmospheric compound by Houzeau (1858), was systematically measured in several places in the late 19th and early 20th centuries including at the Observatory of the Parc Montsouris in Paris (Lévy, 1879; 1907). Half a century later, Arie Haagen-Smit (1952) explained that the frequent health threatening summertime surface ozone episodes (photochemical *smog*) in the Los Angeles Valley were caused by the nitrogen oxides and hydrocarbons emitted by industrial activity and road traffic. These studies highlighted the complexity of atmospheric photochemical mechanisms and the difficulty of developing simple air pollution mitigation measures. Deadly winter air pollution episodes in industrial basins (e.g., Meuse Valley, Belgium in 1930; St Louis, MO in 1939; Dorona, PA in 1948; and London, UK in 1952) were attributed to high aerosol loading resulting from the emission of soot and sulfur oxides during coal combustion.

In the late 1960's, the scientific community became alarmed by the acidification of precipitation and the related detrimental impacts on crops, forests, and aquatic ecosystems. In the 1980's, the focus moved to understanding the oxidizing power of the atmosphere. Since then, much work has focused on understanding the formation and fate of secondary products resulting from the photooxidation of primary pollutants. A large number of airborne field campaigns took place to address fundamental questions related to global and regional aspects of atmospheric chemistry. Space observations started daily global monitoring of air pollutants from the 2000s. Simultaneous advances in supercomputing facilities allowed development of complex chemical models to analyze field observations, perform short- and long-term future projections of air quality, estimate climate forcing and assess the impacts of air pollution on human health, crops, and physical weather and climate processes. Models have become key tools to investigate scientific questions related to chemical weather and chemical climate and very recently have been used to produce daily air quality forecasts in various parts of the world such as the US (Lee et al., 2017), Europe (Baklanov, 2017), China (Brasseur et al., 2019), and India (Kumar et al., 2020a).

3. Chemical Weather

Advances in fundamental knowledge and the availability of frontier technologies to observe and simulate the atmosphere have enabled development of comprehensive chemical weather prediction systems at the global, regional, and local scales. The methodology bears a lot of resemblance with the meteorological forecast's methodology. In both cases, models use similar numerical techniques to solve conservation equations with initial and boundary conditions representative of observed conditions and require near-real-time access to observations from operational monitoring networks including satellites. As for meteorological weather, chemical weather models produce large amounts of data that must be carefully analyzed and evaluated before dissemination to the public. Scoring methodologies to measure the success of forecasts are similar in both types of operations. Advances in data assimilation techniques, initially developed by the meteorology community have improved our skills to forecast air quality (Kumar et al., 2020a).

The approaches used by the two disciplines also have substantial differences. First, meteorological models solve a few (7) equations expressing the momentum, energy, and mass conservation supplemented by the equation of state, the water conservation equation and parameterizations of sub-scale physical and hydrological processes, while chemical models solve a large number (~200-300) of coupled nonlinear continuity equations and require input emissions. The computational burden of models with detailed chemical mechanisms can supersede manyfold that of meteorological models. Second, atmospheric chemists and meteorologists often have different emphasis. Extreme and potentially dangerous weather events are characterized by strong winds, convective situations, heavy precipitations, cyclones, hurricanes, and flooding; while heavy air pollution events are characterized by stable boundary layers, calm winds, and lack of convective motions. Table 1 details the differences between the foci of the two research communities.

Table 1. Comparison between foci of meteorological weather and chemical weather

Variable or emphasis	Meteorological Weather	Chemical Weather
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Temperature	Horizontal and vertical gradients, vertical stability. Ultimate driver of atmospheric circulation	Chemical kinetics, Gas particle partitioning, biogenic VOC emissions, ozone formation
Pressure	Weather patterns, cyclogenesis, local flows (sea breeze, and terrain-induced circulation)	Total air concentration, rates of termolecular reactions
Wind velocity and direction	Transport of heat and momentum, movement of weather disturbances; wind shear: driving force of mechanical turbulence	Multi-scale horizontal transport, dilution of emissions, sea-salt, dust and biospheric emissions;; source-receptor relationship, attribution of pollution sources
Evaporation and precipitation	Latent heat exchange, source and sink of water vapor, ground water content	Removal of water-soluble species and aerosols, fertilization of ecosystems.
Atmospheric humidity	Water cycle, formation of clouds and precipitation	Source of hydroxyl radical and atmospheric oxidation processes
PBL height, turbulence and convection	Vertical exchange of sensible heat, formation of clouds and thunderstorms	Vertical exchange of near-surface chemical species, small-scale dispersion of chemical species, NO _x production in thunderstorms, ventilation of emissions, exchange of chemicals with the free troposphere
Aerosol and cloud microphysics	Scattering, reflection of light; radiative balance, precipitation	Heterogeneous chemistry, photolysis, air quality and health impacts, visibility
Solar radiation	Atmospheric heating, surface absorption, direct and indirect change in energy budget	Photolysis of molecules, ionization of atoms and molecules
Terrestrial radiation	Trapping of infrared radiation by greenhouse gases	Trapping of infrared radiation by aerosols
Land cover	Surface albedo; sensible and latent heat flux, momentum flux, generation of turbulence (roughness)	Surface deposition and biogenic emissions
Surface humidity	Latent heat flux, PBL development, cloud formation, wildfires	Soil emissions, fire emissions
Surface emissions of greenhouse gases and reactive species	Greenhouse effect; methane released from melting permafrost	Source of primary reactive species and formation of secondary species including ozone, air pollution and health impacts;
Extreme events	High winds, heavy precipitation, convective activity, thunderstorms, hurricanes and typhoons, drought, extreme heat events; reduced visibility.	High air pollution events in the stable PBL resulting from high emissions, low wind velocities, lack of precipitation; reduced visibility

Despite the aforementioned similarities and differences, the importance of the two-way interactions between meteorological and chemical processes must be recognized. These include the feedback between the aerosols and the PBL, which prolongs the air pollution episodes (Gao et al., 2015; Ding et al., 2016; Li et al., 2017; Miao et al., 2019). Further, anthropogenic aerosols

may reinforce the strength of hurricanes including lightning rates and precipitation intensities by affecting cloud microphysics and thermodynamics (Pan et al., 2020).

The predictability of coupled meteorological and chemical forecast systems is always limited to a few days due to nonlinear behavior (Brunner et al., 2015). Forecast systems should therefore employ an *ensemble* approach with different realizations. Meteorological weather forecasts represent a classic initial value problem and generally use an ensemble created by slightly perturbing initial conditions in a single model. The chemical forecasts are also affected by external forcing including surface emissions, meteorology dependent chemical kinetics and atmospheric transport; it is therefore preferable to include in the ensemble several independent models. The difference between individual models is usually larger than the difference between the different realizations produced by a single model (Brunner et al., 2015).

Multi-model ensemble based composite chemical forecasts generally perform better than the individual models (Delle Monache and Stull, 2003) due to partial averaging out of the random errors associated with each model. Further, the ensemble approach allows characterization of the robustness of the forecast via investigation of the uncertainty space associated with errors in both the input parameters and model formulation. However, this approach does not necessarily deliver an accurate probability density function of the predicted fields because the models are rarely completely independent and the number of ensemble members is often too small. There is no established evidence that the ensemble mean/median is closer to the true state than the individual model prediction (Mallet and Sportisse, 2006). Figure 1 shows an example in which the median of ozone forecasts provided by eight models represents less accurately the true values (measurements) than the forecast produced by one particular model included in the ensemble (CHIMERE model in this particular case). However, the ensemble approach can be improved by attributing a weight to each model that is determined by an optimization procedure like a least-square error minimization of the ensemble forecasts during a training period covering a few weeks (Pagowski et al., 2005) or via ensemble-calibration using the variance-deficit and model output statistics methods (Kumar et al., 2020b). Large computational costs of a multi-model ensemble can make their implementation in operations challenging but this problem can be

partially addressed using an analog-based method of generating air quality ensemble (Delle Monache et al., 2020).

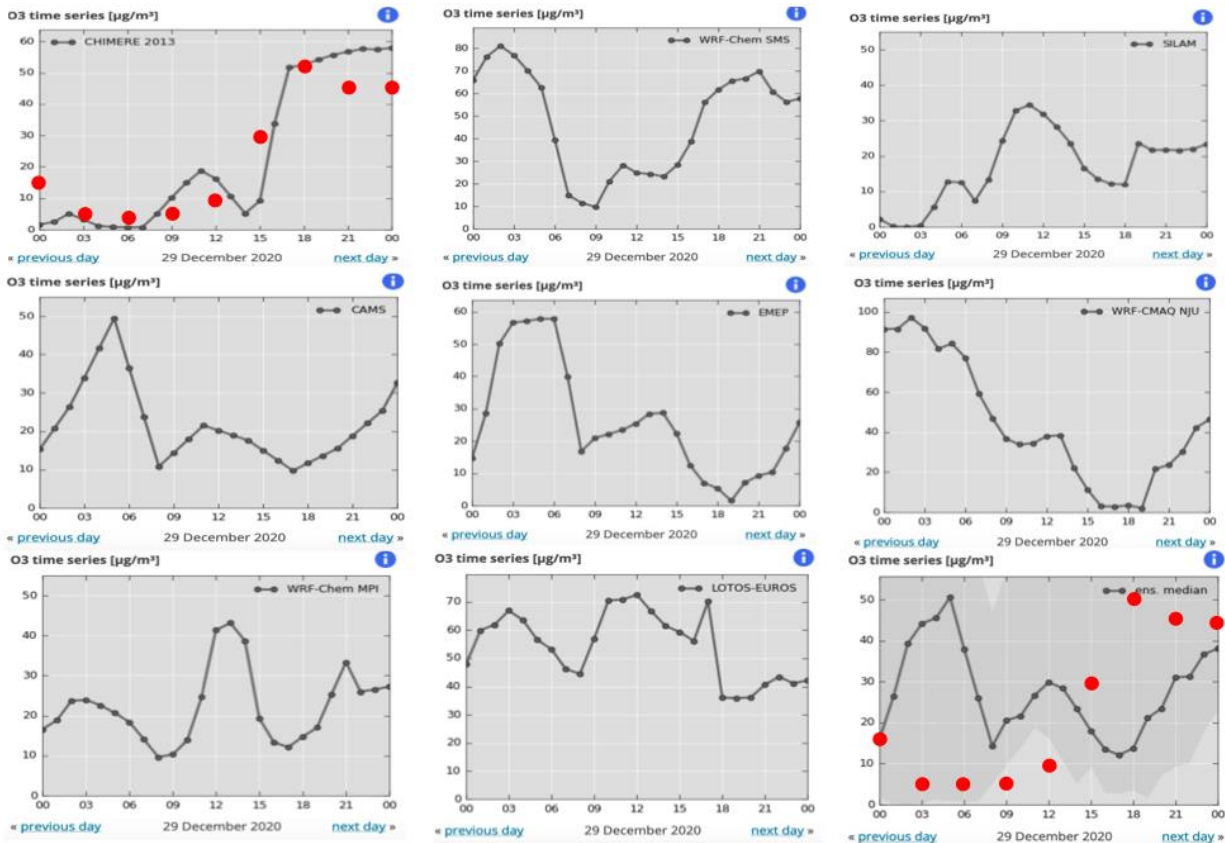


Figure 1. Operational forecast by the MarcoPolo Panda system (Brasseur et al., 2019) of near-surface ozone concentrations (micrograms per m³) in Shanghai, China provided by 8 different chemical meteorological models (CHIMERE, WRF-Chem-SMS, SILAM, CAMS, EMEP, WRF-CMAQ-NJU, WRF-Chem-MPI, and LOTOS-EUROS) for 29 December 2020. The lower panel on the right side provides the median values of the ensemble. The grey shaded area shows the dispersion of the forecasts and the red dots ozone measurements at the US consulate in Shanghai as reported by IQ Air (www.iqair.com/us/China/Shanghai).

4. Chemical Climate

In most applications, the long-term evolution of the chemical composition of the atmosphere is estimated by a single atmospheric model, which is forced by a range of projected emission scenarios such as the Representative Concentration Pathways (RCPs) and Shared Socioeconomic Pathways (SSPs) and, in certain cases, accounts for two-way feedbacks of between chemical and physical climates. As stated earlier, however, it is advisable to take into account the chaotic

nature of the climate system associated with the natural variability in the dynamical state of the atmosphere, (Garcia-Menendez et al., 2017; Pienkosz et al., 2019) and therefore to perform ensemble simulations. Each ensemble member will associate a particular realization of the future chemical composition to a projected state of the climate system. In addition to future projections of the average evolution of the chemical composition, multi-model ensemble simulations provide important information on interannual variability, the amplitude of which is often of the same order of magnitude as that of the average changes. Probability density functions associated with the future state of the atmosphere allows a better differentiation between the effects anthropogenic effects and natural variability with beneficial consequences for the development mitigation measures and environmental policies needed to improve future air quality.

5. Outlook

Predicting chemical weather and chemical climate are more necessary than ever in light of continuing global increase in anthropogenic emissions, recent and projected increase in wildfire activity, and increasing trends of dust aerosols in several parts of the world. This is also recognized in a recent US National Academy of Sciences report, which states that an ultimate goal of atmospheric chemistry research is to establish an effective predictive capability (NAS, 2016). A comprehensive prediction system must include five key components (Kumar et al., 2018): (1) monitoring (in situ and space observations); (2) modeling (multi-scale forecasts); (3) interpretation (evaluation of impacts); (4) dissemination (translation of technical details into actionable information), and (5) training and education (of local students and scientists specifically in the developing world).

Expanding meteorological models to include prognostic atmospheric chemistry (including aerosols) modules is often computationally expensive, although some operational centers are moving into this direction and provide comprehensive environmental predictions (Inness et al., 2019). New methodologies based on artificial intelligence algorithms trained by ensembles of model simulations should be developed to treat atmospheric chemistry more efficiently in large and complex meteorological models. Inverse modeling techniques should be employed to

identify locally and regionally the major sources of pollutants and greenhouse gases, and to develop appropriate and effective mitigation measures that will improve air quality and protect climate.

Developing a predictive capability for chemical weather and chemical climate should be approached in an integrated and holistic perspective, taking into account important dynamical feedbacks for the multi-scale planetary dynamics. As the NAS (2016) report states: “Attaining such a predictive capability requires an understanding of the Earth system developed from laboratory and theoretical studies of fundamental atmospheric chemistry and physics, instruments and atmospheric observations to establish atmospheric constituents and processes, and computational models to integrate understanding. This integration allows attribution of causes of an observed societal impact to particular societal choices.”

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