



Meteorology and Climate Influences on Tropospheric Ozone: a Review of Natural Sources, Chemistry, and Transport Patterns

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Abstract

Tropospheric ozone is a key air pollutant and greenhouse gas. Its fate strongly depends on meteorological conditions and therefore subject to climate change influences. Such dependences through biogenic, chemical, and dynamic processes on different spatiotemporal scales have been unraveled from observations and modeling studies. In this process-oriented review, we summarize three dominant pathways of meteorological and climatic impacts on tropospheric ozone and present their recent progress. The three pathways are influences through changes in the natural precursor emissions, the kinetics and partitioning of chemistry and deposition, and the transport of ozone and its precursors. Tropospheric ozone levels have shown significant global or regional responses to meteorological/climatic changes (e.g., changes in the Brewer-Dobson Circulation, the Hadley Circulation, and El Niño–Southern Oscillation) and can be explained through the conjunction of these pathways. Most recent model projections predict that future climate will increase surface ozone in polluted regions and decrease ozone at a global scale due to stronger ozone chemical loss. However, uncertainties in climate-ozone responses and limitations in model capability still challenge the magnitude and even the sign of such projections. We highlight the rising importance of future increase of stratosphere-troposphere exchange in modulating tropospheric ozone that may largely compensate the predicted chemical loss of tropospheric ozone burden. We also highlight that uncertainties in isoprene chemistry, biogenic emissions in changing CO₂ levels and vegetation, and interactions between ozone and vegetation may largely affect the surface ozone response to climate change. Future research and model improvements are required to fill these gaps.

Keywords Tropospheric ozone · Ozone · Meteorology · Climate · Natural sources

Introduction

Ozone at the surface is detrimental to human health and ecosystem [123], while in the middle and upper troposphere, it is a greenhouse gas contributing to positive radiative forcing [175, 184]. Efforts of reducing anthropogenic emissions of ozone precursors such as nitrogen oxides (NO_x = NO + NO₂) have been applied to improve ozone air quality particularly in

Europe and North America [51]. However, as the natural sources, chemistry, and transport of ozone and its precursors are highly climate-sensitive, the effectiveness of such efforts will be modulated by climate variations or even offset by unfavorable weather conditions, imposing challenges for ozone quality management. As such, it is of particular importance to evaluate the connections between tropospheric ozone and meteorological conditions (and associated climate variations), and their implications for future ozone projection in the context of climate change. We review our current understandings and recent advances on this issue.

Meteorology variations and climate change influence tropospheric ozone through a number of processes. We summarize three dominant pathways in Fig. 1, including (1) natural emission pathway, i.e., a large amount of ozone precursors are emitted from climate-sensitive natural sources such as lightning and biosphere; (2) chemistry pathway, i.e., meteorological conditions such as solar radiation, temperature, and humidity alter the partitioning and efficiency of chemical reactions

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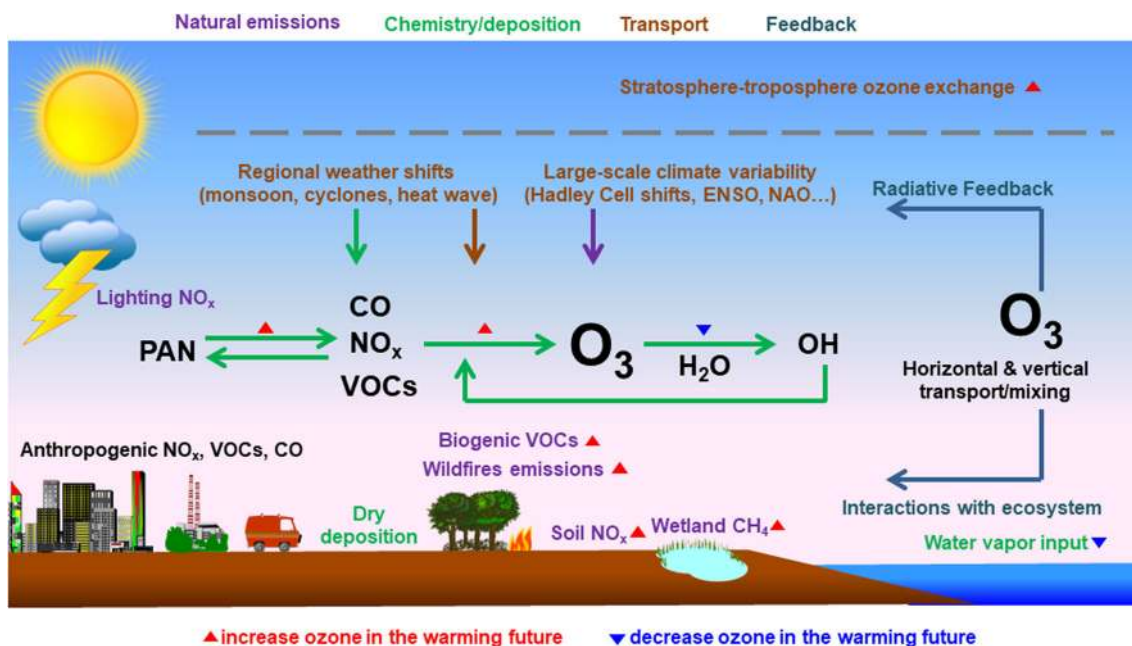


Fig. 1 Pathways of interaction between meteorology/climate changes and tropospheric ozone. The red (blue) triangles represent that at global scale future climate will increase (decrease) tropospheric ozone through

the specific pathway based on current understanding. More discussions are provided in the text

and dry deposition, and therefore modulate ozone production and loss; and (3) transport pathway, as the lifetime of ozone and its precursors in the free troposphere can be longer than months, they are subject to changes of transport patterns on different spatiotemporal scales. It shall be noted that the impacts of meteorology and climate on tropospheric ozone often appear as a conjunction of more than one pathway. Tropospheric ozone changes in turn alter climate through radiative feedback and interactions with the biosphere (Fig. 1).

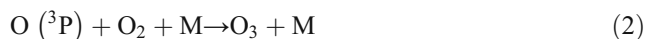
The overall responses of tropospheric ozone to changes of meteorology and climate have been summarized in previous reviews [46, 47, 78, 82, 205]. The responses are generally quantified through observed statistical relationships of ozone with meteorological variables, or through perturbation analyses using chemical models [82]. One distinguished finding is the positive surface ozone-temperature relationship in the polluted regions, mainly driven by the role of temperature in increasing natural emissions (in particular biogenic isoprene emissions) and accelerating ozone chemical production at high NO_x levels [150]. The positive ozone-temperature relationship implies that global warming will deteriorate surface ozone air quality in industrial regions even without increases of anthropogenic emission, an impact referred as “climate penalty” [217]. Previous reviews also documented the relationship between ventilation conditions (stagnations and cyclones) and ozone air quality, and summarized future ozone projections driven by climate change, although the confidence of such projections can be limited by uncertainties in chemical mechanisms (such as organic nitrates production) and the lack of atmosphere-biosphere interactions in the model [46].

Different from previous reviews which focus on the overall ozone response to climate change, this study aims to present a process-oriented review on how meteorology and tropospheric ozone interacts through each of the pathways. A number of recent progresses of these processes are also included. Particularly, recent studies have shown that shifts of stratosphere-troposphere exchange (STE) and large-scale climate patterns such as the El Niño–Southern Oscillation (ENSO) and Atlantic Multidecadal Oscillation (AMO) have significant impacts on present-day ozone distribution and future ozone projections. We include these important responses in the review. The review is organized as follows. The three pathways as described in Fig. 1 are reviewed in the “Effect on Natural Sources of Ozone Precursors” section, the “Effect on Ozone Chemistry and Deposition” section, and the “Effect on Ozone and Precursor Transport Patterns (Associated with Weather and Climate Patterns)” section, respectively. We summarize recent studies (since 2009) of future tropospheric ozone projections due to climate change in the “Future Ozone Change Due to Climate Change” section, and discuss the feedback from tropospheric ozone to climate in the “Feedback from Tropospheric Ozone Change to Climate” section. A conclusion is provided in the “Conclusion” section.

Effect on Natural Sources of Ozone Precursors

We start with a brief overview on tropospheric ozone chemistry summarized from Jacob [81], Atkinson [7], and Wang et al.

[210]. In the troposphere, photolysis of NO_2 (at wavelengths < 424 nm) provides $\text{O}(^3\text{P})$ (the ground electronic state oxygen atom) (1). Ozone is then formed through a termolecular reaction of $\text{O}(^3\text{P})$, O_2 , and a third body M (2).



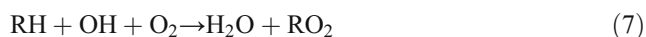
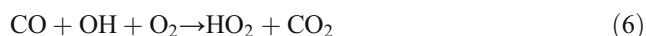
O_3 reacts rapidly with NO to regenerate NO_2 through (3),



contributing to null ozone production through (1)–(3). However, the presence of oxidant radicals (hydroperoxyl radical (HO_2) and organic peroxy radicals (RO_2)) provides additional pathways to convert NO to NO_2 through (4) and (5),



RO_2 , HO_2 are products from oxidation of CO (6), hydrocarbons (RH, 7), or alkoxy radicals (RO) (8).



The oxidation of CO and hydrocarbons requires hydroxyl radical (OH). It originates principally from photolysis of O_3 (9) and reaction with water vapor (10).



The above mechanisms present the tropospheric ozone production through a chain photochemical oxidations of CO and hydrocarbons (or in broader context, volatile organic compounds (VOCs)) catalyzed by HO_x ($\text{HO}_x = \text{OH} + \text{H} + \text{peroxy radicals}$) in the presence of NO_x . The chain is terminated by the loss of HO_x radicals, which happens through the oxidation of NO_2 by OH (11), and the self-reaction of HO_2 (12):



H_2O_2 and HNO_3 are then removed mainly by wet deposition due to their high solubility in water.

Ozone chemistry is strongly nonlinear. At low NO_x levels, the controlling termination is (12); thus, ozone production is limited by the supply of NO_x and is not sensitive to hydrocarbons, referred as “ NO_x -limited regime.” At high NO_x levels, the controlling termination is (11); thus, ozone production linearly increases with VOCs concentrations but decreases with NO_x concentrations, referred as “VOC-limited regime.”

Meteorological conditions therefore influence ozone through modulating the climate-sensitive natural emission of its precursors, including soil NO_x emissions (“Soil NO_x Emission” section), lightning NO_x emissions (“Lightning NO_x Emission” section), biogenic VOC (BVOC) emissions (“BVOC Emissions” section), wildfire emissions (“Wildfire Emission” section), and wetland methane emissions (“Wetland Methane Emissions” section). We present a process-based description on the role of meteorology in each process and discuss the ozone response.

Soil NO_x Emission

NO_x can be produced naturally from soil as byproduct of microbial activities (nitrification and denitrification). Soil emissions contribute to approximately 10–15% (3 to 8 Tg N year⁻¹) of the present-day global NO_x emissions ([31, 77, 196, 204]). It is controlled by inorganic nitrogen availability in soil, canopy structure (such as vegetation type), and edaphic conditions such as soil temperature and moisture [204, 226].

Soil temperature and moisture are critical factors in controlling soil NO_x emissions. These two factors together can explain up to 74% of the observed variations of soil NO_x emissions in European forests [160]. Rising soil temperature accelerates the enzymatic process and exponentially increases soil emissions as observed over different biomes [160, 222]. The dependence of soil emissions on temperature, however, weakens when soil temperature further increases (e.g., above 30 °C) and soil becomes dry, and then soil emissions become more limited by water content [222]. High soil moisture also suppresses soil NO_x emissions, as wet condition with fewer oxygen supply favors denitrification which preferentially emits N_2O and N_2 , and also limits gas diffusion through the soil pores [77, 222]. Further impacts from soil moisture can be found when there is a sudden shift from dry to wet conditions. The sudden shift can release accumulated inorganic N rapidly and reactivate the water-stressed bacteria, leading to a burst of soil NO_x emission. Rapid and intense soil NO_x pulsing emissions following rainfall in the US and India have been captured by daily satellite NO_2 observations [13, 53].

Changes in soil NO_x emissions due to variations of meteorology and climate further modulate ozone. As soil emissions dominate in rural regions where ozone chemical production is typically NO_x -limited, it is expected that soil NO_x emissions trigger strong local ozone production. Romer et al. [152] showed that soil NO_x emissions contributed to nearly half of the ozone increases with rising temperature in a rural site in the southeastern US. Hudman et al. [76] showed that warmer (2 K) and drier (50%) weather conditions followed by convective precipitation over the central US in June 2006 led to about 50% higher soil NO_x emissions compared to the average for 2005–2008, mainly due to stronger pulsing emissions in

that year. Increased soil NO_x emissions alone then led to surface ozone enhanced by 3–5 ppbv (episodically up to 16 ppbv). Similar enhancements (May–August 2017 vs. 2016) of soil NO_x emissions (~25%) and surface ozone (1–2 ppbv) due to warmer climate were simulated over the industrial eastern China [119]. For future projections, modeling studies predict significant enhancement of soil NO_x emissions driven by climate change (e.g., ~23% higher in 2100 compared to 2000 in IPCC A2 emission scenario, [61, 108]), underlying future climate will likely degrade ozone air quality via increasing soil NO_x emissions.

Lightning NO_x Emission

Energy produced by lightning flashes dissociates and converts atmospheric N₂ molecules into NO_x. Estimated global lightning NO_x emissions are ranging from 2 to 8 Tg a⁻¹ N with large uncertainties [31, 126, 163]. The importance of lightning NO_x in atmospheric chemistry and potential radiative effect is disproportionally large as it is mainly released in the upper troposphere, where ozone chemical production is more efficient, and where NO_x and ozone have longer lifetimes [10].

Lightning NO_x emissions strongly depend on the intensity and frequency of lightning activities in the convective thunderstorms. Price and Rind [149] showed that the total lightning flash frequency in the thunderstorm exponentially increased with convective cloud top height (CTH) with a power of 4.9 in continental cloud. Several studies also linked lightning flashes to other convection-related characters such as updraft velocity, latent heat release, and more recently upward cloud ice flux [40, 42, 163]. These dependencies are then parameterized into models to estimate lightning NO_x emission and ozone production. Lightning emissions contribute to upper tropospheric ozone by more than 10 ppbv [27, 75], and also influence surface ozone especially at regions with high elevations such as the US Intermountain West and the Tibetan Plateau [119, 238]. It is also an important driver of observed interannual variability of ozone and OH in tropical upper troposphere [125, 127].

Climate variabilities can then influence tropospheric ozone through altering lightning NO_x emissions. Anomalous, high ozone contributed by lightning emissions in El Niño conditions (“Large-Scale Climate Patterns (ENSO, AMO, NAO)” section) has been found at tropical upper-troposphere [55, 130] due to intensified convection over land and coastal area [58]. The projected changes of future lightning ozone productions due to climate change, however, largely depend on the parameterization of lightning in the model. Most studies with lightning parameterized based on CTH showed enhancements of lightning NO_x emissions (4–60% K⁻¹) in the warming future due to more frequent and intense convections [163]. However, studies that used cloud ice flux for parameterization resulted in an opposite conclusion, as the cloud ice crystal

declines with increasing temperature [41, 42, 83]. Therefore, the projections of future lightning and its impact on ozone need to be interpreted with caution.

BVOC Emissions

VOCs are important ozone precursors, a large amount of which are emitted from terrestrial ecosystems. BVOC emissions vary among plant functional types and are strongly modulated by meteorological conditions. Temperature is one of the key factors controlling BVOCs emissions due to the nature of photosynthesis. Exponential enhancements of biogenic isoprene and monoterpene emissions with rising temperature have been shown in field and laboratory observations and implemented in chemical models [57]. The exponential dependency of BVOC emissions on increasing temperature is also identified as a main driver of the positive ozone-temperature correlations especially over urban areas where NO_x levels are high [119, 150]. Modeling results showed that a 3 K temperature enhancement on BVOC emissions alone would increase biogenic isoprene emissions by 6–31% and surface ozone by > 2 ppbv in the northern mid-latitudes [35]. The increased isoprene also affects the partitioning among oxidized nitrogen to produce more peroxyacetyl nitrate (PAN, a NO_x reservoir compound), which can transport a long distance and produce ozone downwind ([45]; see also the “HO_x Chemistry” section).

BVOC emissions are suppressed at extreme high temperature conditions (e.g., > 40 °C) which adversely affect cellular activities [56]. The suppression of biogenic isoprene emissions can explain the observed decline of surface ozone at extreme high temperatures (> 312 K) over California [183]. Drought conditions also impede isoprene emissions as decreasing water content slows down photosynthetic rate and stomatal conductance. Jiang et al. [87] estimated that including the drought effect in the model would lead to reduction of biogenic isoprene emissions by 17% globally. However, there is evidence that in the initial phase of drought, the shutdown of the plant physiological processes can enhance BVOC emissions [87, 144, 148]. Zhang and Wang [235] showed that enhanced biogenic isoprene emissions from water-stressed plants at the onset stage of drought contributed to the abnormally high ozone episodes over the southeast US in October 2010.

Model projections tend to predict significant increases of BVOC emissions in the warming future, which would elevate tropospheric ozone concentrations (e.g., [109, 199, 217]). However, these projections might be influenced by uncertainties in isoprene chemistry and interactions with the biosphere as pointed out by recent studies and summarized as follows:

1. Uncertainties in isoprene chemistry. Whereas oxidation of the emitted BVOCs by OH produces RO₂ (7) and further

generates NO₂ (5) and ozone, RO₂ and NO can also go through another branch that forms isoprene nitrates (RONO₂),

$$\text{NO} + \text{RO}_2 + \text{M} \rightarrow \text{RONO}_2 + \text{M} \quad (13)$$

RONO₂ presents as a sink of both NO_x and RO₂, thus inhibits ozone production. The ratio of (13) branch in the total (NO+RO₂) reaction is estimated to be 10 ± 5% [142, 219], depending on a variety of factors including temperature [7, 164]. Isoprene nitrates could be either recycled to regenerate NO₂ and ozone, or be deposited to surface [142]. Therefore, different chemical mechanisms of isoprene oxidations (whether include (13) or not, include recycle or not, and their ratios) presented in the models determine the sensitivity of ozone to perturbed temperature and biogenic isoprene emissions [49, 79, 182, 216]. Through modeling studies, Ito et al. [79] showed that if no RONO₂ were recycled to NO_x, the ozone burden would be 17 Tg higher in a 5 K warmer scenario than the cooler scenario, while if all RONO₂ were recycled, ozone burden differences between the two scenarios would be much larger (57 Tg). Fu et al. [49] also showed that assuming a higher cycling rate of RONO₂ (55% versus 0%) in the model produced a larger sensitivity of surface ozone to temperature (8 ppbv K⁻¹ vs. 5 ppbv K⁻¹ in 2000) in the southeastern US. Improving understanding of isoprene chemistry mechanism is therefore critical for estimating the climate-BVOC-ozone response.

2. Response to future ambient CO₂ concentrations. Laboratory and field observations have shown substantial reductions in isoprene synthesis at elevated ambient CO₂ levels [6]. As such, future CO₂ increases could largely offset [63, 191], or even counteract the warming induced enhancements of BVOC emissions [59]. Tai et al. [191] showed that including CO₂ inhibition on BVOC emissions in the model decreased projected future surface ozone in eastern US, Southeast Asia, and Europe by 6 ppbv compared to the results without CO₂ inhibition, but increased ozone in western Amazon, central Africa, and Southeast Asia, where reduced sequestration of NO_x by isoprene oxidation products enhanced NO_x levels in these NO_x-limited regions [228]. The above studies all point to the important role of the CO₂ inhibition effect on BVOC emissions that may change the magnitudes and signs of future ozone projections, yet most of the current projections tend to miss this mechanism in the models (see also “Future Ozone Change Due to Climate Change” section).

3. Changes in land cover/vegetation types. Future environment (e.g., higher CO₂ fertilization, changes in temperature and precipitation) could naturally alter the abundance and distribution of vegetation, which may lead to large discrepancies in the projected effect on BVOC emissions. Sanderson et al. [158] showed that the climate-driven changes of vegetation types (e.g., the recession of tropical forests) would lead to less BVOC emissions, while Wu et al. [218] found increases in

global isoprene emissions. More recently, Hantson et al. [59] found that such different responses largely depended on the relative changes of different plant functional types.

Wildfire Emission

Wildfires emit large amounts of CO, NO_x, and VOCs and produce approximately 170 Tg year⁻¹ (about 3.5% of the annual total chemical production) of ozone with large interannual variability [84]. Meteorology can alter wildfire emissions and associated ozone production through modulating (1) wildfire frequency and intensity, (2) emitted tracers, and (3) ozone photochemistry in wildfire plumes.

Wildfires are prone to occur in hot and dry weather conditions. The intensity and frequency of wildfires have been increasing in the western US since 1970 due to rising temperature and earlier snowmelt [214]. Lu et al. [116] estimated the relationship between meteorological parameters and summertime wildfire frequency and intensity at monitoring sites in the western US. They found that occurrences of large wildfire events could enhance notably with increasing temperature and solar radiation, and with decreasing relative humidity and wind speed. When temperature was higher than 30 °C, the frequency of large wildfire events was four times higher than that of small events. Wildfire emissions are also influenced by combustion efficiency, which largely depends on meteorological conditions [168]. High temperature favors flaming combustion (high combustion efficiency), leading to stronger oxidation of fuel nitrogen compounds, larger proportion of NO_x emissions, and therefore higher ozone production. Smoldering combustions in cooler conditions, on the other hand, tend to release higher proportion of reduced nitrogen compounds such as NH₃ and are not favorable for ozone production [84].

Ozone chemical production in wildfire plumes is also subject to meteorological conditions. Low temperature typically in boreal wildfires favors rapid conversion from emitted NO_x to PAN. It limits ozone production near fire burning spots but may lead to ozone enhancement downwind ([3], see also “PAN Chemistry” section). Vertical diffusion influences the injection heights of wildfire plumes which are critical to ozone production and transport [200, 246]. At higher altitudes, the wildfire plumes are exposed to higher solar radiation without the blocking of wildfire aerosols and can also be more efficiently transported downwind [86, 141]. All these complexities in meteorology-relevant wildfire emissions and chemistry lead to a wide range of observed wildfire ozone enhancements as the plumes travel and age ([84], and reference therein), and make it difficult for chemical transport models to capture wildfire ozone influences especially at coarse grid resolution [116, 238].

Hot and dry weather condition then favors wildfire ozone enhancement, as it increases the frequency and intensity of

wildfire, enhances the combustion efficiency, and facilitates wildfire ozone chemical production. Summertime wildfire ozone enhancements in the western US could be 1–3 ppbv higher in hot and dry years such as 2002–2003 than other years [116]. Predictions of future wildfire activities have been available in several climate models or vegetation models [93, 203], all suggesting increasing burned area and wildfire emissions in the warming future, consistent with previous projections based on statistical methods [19, 181, 230].

Wetland Methane Emissions

Methane is an important ozone precursor in remote regions due to its long chemical lifetime (about 9 years). Wetland emissions (100–250 Tg year⁻¹) are the dominant natural source accounting for 20–50% of the total methane emissions [92, 159]. Wetland releases methane when bacteria reduce organic carbon to methane under the anaerobic environment [15]. This process is controlled by soil temperature which influences bacteria activity, water table position which determines production and oxidation depth, carbon availability (soil carbon substrate), and decomposition rate [14, 159]. Increasing temperature accelerates the methane production and oxidation rates. Increasing precipitation extends wetland areas and raises water tables; both enhance wetland methane emissions [134]. Christensen et al. [20] showed that soil temperature explained 84% of the methane emission variations over a number of northern wetland sites. Recent studies pointed out that climate variabilities such as ENSO could partly explain the interannual variations of wetland methane emissions especially in tropics through changes in temperature and precipitation [68, 245].

Significant enhancements of wetland methane emissions are projected with future increases in temperature and precipitation [134], although the enhancements may be partly offset by the effect of soil moisture depletion [18]. Shindell et al. [174] showed that global wetland methane emissions would increase by 78% if CO₂ concentrations double in the future. Increasing wetland methane emissions would cause a cascade of chemical influences and climate feedbacks. It could enhance ozone concentration, influence global OH burden [174], amplify methane chemical lifetime, exert a strong radiative forcing that faster the warming [52], and further increase methane emissions from wetland and thawing permafrost [134].

However, so far, only few models include interactive climate-sensitive wetland methane emissions, with the majority using prescribed methane mixing ratios for the future ozone projection [99, 124]. To our knowledge, future ozone changes due to increasing wetland methane emissions have not been comprehensively quantified so far. Our current understanding of ozone production from climate-sensitive natural methane sources such as permafrost, lakes and ponds

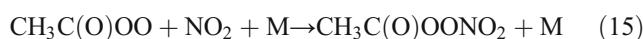
[215], and marine methane hydrate [153] are rather limited, and should be addressed in the future studies.

Effect on Ozone Chemistry and Deposition

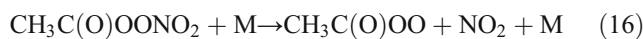
Meteorology can influence tropospheric ozone through modulating the rate of chemical kinetics, the partitioning of reaction pathways, and efficiency of deposition. In this session, we discuss changes in ozone production and loss due to climate-sensitive PAN chemistry (“PAN Chemistry” section), HO_x chemistry (“HO_x Chemistry” section), and dry deposition (“Dry Deposition” section).

PAN Chemistry

PAN is generated through the oxidation of acetaldehyde in the presence of NO_x in hydrocarbon-rich environment ((14) and (15)) [81].



It is removed mainly via thermal decomposition (16) in the lower troposphere below ~7 km [192].



One important feature of PAN is the dependence of its lifetime on temperature. Because the decomposition rate of PAN (16) drops dramatically with decreasing temperature, the lifetime of PAN extends from 30 min at 295 K to several months at 240 K [7, 81]. This feature allows temperature to influence the production and transport of ozone through PAN chemistry. The formation of PAN serves as sinks for both NO_x and peroxy radicals, and therefore lowers ozone production near the source region. Nevertheless, PAN can be transported to a long distance in the cold free troposphere, eventually be thermally decomposed to release NO_x (often due to air heating with subsidence), and consequently enhances ozone production with high efficiency in remote regions [48]. Previous studies have shown the role of PAN as a NO_x reservoir compound that helps transport NO_x from polluted regions such as east Asia [236] and fire spots [3] to remote regions and produce ozone there. Increasing PAN thermal decomposition with rising temperature is also a driver of the observed positive ozone-temperature correlation [150], but this relationship could be much weaker at extremely high temperature (e.g., > 312 K, [183]).

Temperature increases in the future will lead to stronger thermal decomposition on PAN, resulting in ozone increases in the polluted regions but decreases in remote regions. Doherty et al. [35] showed that a 3 K temperature increase

on the chemical reaction rate coefficient of (16) would cause up to 4.2 ppbv ozone enhancement over land and up to 1 ppbv ozone decrease over the remote oceans. The decrease of PAN over remote regions, however, can be compensated by increasing PAN generated from higher BVOC emissions in warmer climate as discussed in the “BVOC Emissions” section.

HO_x Chemistry

Atmospheric water vapor (HO_x sources) is essential to ozone photochemistry. Its influences on tropospheric ozone are sensitive to ozone chemical regimes. In the remote regions where NO_x levels are low, ozone removal by HO_x is effective, resulting in significant negative correlations between ozone concentration and relative humidity (as a proxy of HO_x concentration), e.g., ozone vs. relative humidity correlation of -0.69 in the summertime western US in 1989–2010 [116]. In polluted regions where NO_x levels are relatively high, water vapor has competing effects on ozone production. OH radical oxidizes CO and hydrocarbons through (4)–(8) and activates ozone production, while it also terminates ozone formation by converting NO₂ to nitric acid (HNO₃) (11), leading to a more complicated relationship between ozone and water vapor [82]. These weak or sign-varied correlations have been revealed in urban areas in Europe and the US [9, 17].

From a global perspective, increasing water vapor in the warming future would lead to a decline of tropospheric ozone burden [205]. Doherty et al. [35] showed that 19% increase of water vapor would reduce surface mean ozone concentrations by 1–2 ppbv for global average and 3 ppbv in the tropics. In the US, however, Dawson et al. [29] showed that 20% decrease of absolute humidity would reduce the national mean surface ozone by 0.5 ppbv. A positive response of ozone to increasing water vapor content was also found in California in a recent study [69], reflecting the competing role of water vapor in tropospheric ozone in polluted regions.

Dry Deposition

Dry deposition to vegetation is an important sink of tropospheric ozone, accounting for about 20% of the annual total tropospheric ozone chemical production [196]. Ozone dry deposition dominantly occurs over vegetated surfaces via stomatal uptake on leaf surface and nonstomatal uptake on plant canopies [60, 243]. It is typically described separately by three processes: turbulent transport in aerodynamic layer, molecular diffusion through the quasi-laminar boundary layer, and uptake at the surface [213]. These mechanisms are commonly parameterized by analogy to the Ohm's law that considers the deposition resistance (reciprocal of deposition velocity) as

electrical circuits: resistances in aerodynamic layer (R_A), quasi-laminar layer (R_B), and surface resistance (R_C).

Dry deposition is significantly influenced by meteorological conditions such as air stability and soil moisture. Strong air stability results in large R_A and impedes dry deposition. At daytime when turbulent is active (small R_A), ozone dry deposition is usually limited by R_C . R_C is further decomposed into stomatal uptake on leaf surface and nonstomatal uptake on plant canopies and ground, both linked to meteorological conditions. Stomatal ozone uptake is controlled by light that controls stomata activity and is also influenced by soil moisture and relative humidity. Drought and high temperature in air or soil would suppress stomatal uptake (therefore suppress dry deposition) due to the closure of stomata to protect plants from desiccation. This mechanism significantly influences ozone in semi-arid regions such the Mediterranean [4] and helps to explain the negative ozone-humidity correlations in the US [89]. Model results also showed that reductions of ozone dry deposition due to persistent high temperatures and drought could contributed to high ozone levels in Europe [179] and China [119]. The nonstomatal ozone deposition, which describes the thermal decomposition of ozone with external surfaces including soil and canopy, also shows some degrees of dependence on temperature and solar radiation [123]. A recent modeling study showed that the Monin-Obukhov length (a parameter for quantifying air stability) and surface temperature, were respectively, key factors influencing model estimates of ozone dry deposition velocity during nighttime and daytime [241].

To our knowledge, the responses of ozone dry deposition to future climate change have not yet been comprehensively quantified. There is one effort by Andersson and Engardt [5], which found that in winter decreasing snow cover in warmer future climate would lead to more effective ozone dry deposition, while in summer, changes in air stability, soil moisture, and temperature would lead to increase aerodynamic and surface resistances (therefore suppress ozone dry deposition). All these effects together led to ozone enhancements of up to 6 ppbv in Europe. They also found that the weaker dry deposition explained more than 60% of the total ozone enhancements, outweighed the effect from increasing biogenic isoprene emissions, implying the important role of dry deposition in climate-induced future ozone changes.

Effect on Ozone and Precursor Transport Patterns (Associated with Weather and Climate Patterns)

As the lifetime of tropospheric ozone and its precursors (e.g., CO, PAN) can reach weeks or months in the free troposphere [229], it allows shifts of transport patterns (typically associated with weather and climate patterns) to influence

tropospheric ozone by redistributing them. Based on the spatial scales, these weather patterns can be classified as synoptic circulations (~ 1000 km), large-scale climate patterns (~ 10,000 km), and global vertical circulations (e.g., [47, 82]). The physical mechanisms of ozone response to these weather patterns have been documented from ground-based measurements, satellite observations, and modeling studies. This section will focus on a “transport” perspective and also combine with discussions in the “Effect on Natural Sources of Ozone Precursors” and “Effect on Ozone Chemistry and Deposition” sections to illustrate that the responses are often associated with changes in natural emissions and chemistry. We will start with the response of tropospheric ozone to STE (associated with large-scale circulation, “STE and Large-Scale Meridional Circulations” section), to large-scale climate variability (~ 10,000 km) such as ENSO and AMO (“Large-Scale Climate Patterns (ENSO, AMO, NAO)” section), and then changes driven by synoptic circulations (~ 1000 km) such as monsoons, subtropical highs, and mid-latitude jet streams (“Synoptic Patterns” section).

STE and Large-Scale Meridional Circulations

From a global and long-term perspective, STE is driven by the large-scale stratospheric meridional circulation known as the Brewer-Dobson circulation (BDC). BDC is characterized by upwelling from troposphere to stratosphere in the tropics, transport to the extratropical stratosphere, and descending from stratosphere to troposphere at middle and high latitudes [185]. STE also occurs episodically at mid-latitudes associated with synoptic scale and mesoscale processes, such as tropopause folds near the jet streams, gravity wave breaking, and deep convections [185, 193]. The role of STE in modulating tropospheric ozone (550 Tg year^{-1} , approximately 10% of the annual global tropospheric ozone chemical production) and surface ozone has been well documented [67, 73, 178, 195].

BDC has been strengthening and is expected to further intensify in the warming future [16]. Increasing tropospheric greenhouse gases and depletion of polar stratospheric ozone (particularly in the Southern Hemisphere) can intensify meridional temperature gradient in the upper troposphere/lower stratosphere (UTLS) region, which enhances planetary wave activity and strengthens the BDC [16]. It then leads to ozone increase in the mid-latitude lower stratosphere and further descends to the troposphere [38, 187, 234]. Hegglin and Shepherd [65] showed that STE ozone transport would enhance by 23% in 2095 compared to the 1965 conditions due to strengthening BDC in the IPCC A1B scenario. Banerjee et al. [11] showed that future climate change alone would increase STE by 17% and 28% in 2100 compared to 2000 conditions for RCP 4.5 and RCP8.5 scenarios, respectively. A more recent study estimated a larger enhancement of STE by 50% for RCP 8.5 [122]. The implications for future ozone

change will be discussed in details in the “Future Ozone Change Due to Climate Change” section.

Tropospheric ozone is also affected by changes in strength and location of the subtropical jet streams or mid-latitude storm tracks where episodic STE occurs [72, 100]. There is observational evidence that subtropical jet streams and mid-latitude storm tracks have been moving poleward (a feature also diagnosed as widening of the Hadley Circulation/tropical belt) [74, 80, 121, 227] most likely caused by changes in meridional temperature gradients in the UTLS [115, 194]. A recent study by Lu et al. [120] attributed the large-scale positive tropospheric ozone trends in the Southern Hemisphere over 1990–2010 to widening of the Hadley circulation, by demonstrating the resulting changes in transport patterns favored stronger STE and ozone chemical production in the Southern Hemisphere. Positive tropospheric ozone trends at individual sites were also reported and linked to stronger STE (e.g., [112, 139]). Xu et al. [221] showed that increasing STE likely associated with strengthening of the mid-latitude jet stream explained approximately 70% of the observed springtime ozone enhancements at Mt. Waliguan Observatory (3816 m) in western China over 1994–2013. Linkages between STE and climate variabilities such as ENSO and the North Atlantic Oscillation (NAO) have also been reported and will be discussed in the next section.

Large-Scale Climate Patterns (ENSO, AMO, NAO)

ENSO

ENSO is one of the dominant climate models that modulates global climate variability and also influences tropospheric ozone on the interannual timescale. In the El Niño condition, tropospheric ozone decreases (increases) in the eastern (western) Pacific regions, as illustrated by negative (positive) correlations between the Niño 3.4 Index and tropospheric column ozone (TCO) over the Pacific seen from satellite observations and model simulations [136–138, 248]. These responses can be explained by changes in zonal transport patterns. In the El Niño condition, the warm ocean shifts eastward into the coasts adjacent to the South America. Abnormal air upwelling above the warmer water in the eastern Pacific lifts the ozone-poor marine air and lowers TCO. Meanwhile, strengthened subsidence occurs in the western Pacific, increasing ozone concentrations there.

Besides influencing transport pathways, ENSO also affects ozone through altering chemistry and precursor emissions. Abnormal uplift in the eastern Pacific in El Niño brings more water vapor (sources of HO_x) into atmosphere, leading to stronger ozone chemical loss ((9) and (10)). The drier western Pacific is, in contrast, more favorable for ozone production than that in La Niña. Sekiya and Sudo [165] showed that although the impacts from transport outweighed those from

chemistry globally, they were comparable over the central Pacific. Warmer and drier weather conditions in the western Pacific during El Niño also promote biomass burning there [176] and enhance lightning activity as discussed in the “Lightning NO_x Emission” section, both contributing to higher ozone [237]. The response of tropical tropospheric ozone to ENSO therefore well illustrates that climate influences ozone through a conjunction of pathways of natural precursor sources, chemistry, and transport.

While the ozone-ENSO response is most significant in tropics, it can expand to mid-latitudes. The El Niño condition, also characterized as easterly shear Quasi-Biennial Oscillation (QBO) phase [100, 131, 247], can enhance STE at mid-latitudes due to stronger subtropical jet than La Niña [166]. Zeng and Pyle [233] found that STE increased the global tropospheric ozone burden by about 4 Tg following the strong 1997–1998 El Niño event. Regionally, higher TCO (4.9 DU) over the Europe in spring 1998 was found associated with stronger STE, Asian pollution transport, and wildfires [94]. Shifts in the polar stream position after La Niña winter have shown to increase frequency of deep stratospheric ozone intrusion events in the western US [111]. Changes in meteorological conditions and transport patterns in El Niño years have also found to cause surface ozone increases in the eastern US but decreases in the southern and western parts [169, 220]. On a 30-year time scale, Lin et al. [110] found that weaker transport from Eurasia to Mauna Loa (Hawaii) observatory, driven by more frequent occurrence of La-Niña-like conditions from 1980 to 2011, contributed to the flattening of springtime ozone, which offset the ozone enhancement due to increasing anthropogenic emissions.

AMO

On the multi-decadal timescales, AMO exerts considerable influences on the global and regional meteorological variability (e.g., [21]). To our knowledge, only a few studies have examined its influence on ozone air quality [170, 173, 223]. AMO is a climate cycle that features positive sea surface temperature (SST) anomalies in the northern Atlantic in its warm phase. Since 1900s, there have been warm AMO phases over 1931–1960 and 1990–2012 and cold phases in 1900–1929 and 1960–1994 [173, 189]. In the warm phase, warming Atlantic SSTs can trigger diabatic heating in the atmosphere, which further influences the extratropical climate through stationary wave propagations [189, 190]. This results in hotter, drier, and more stagnant weather in the eastern US and favors high ozone concentrations there. Understanding such linkages between ozone and SST [225] is particularly valuable because sea heat content has longer memory than atmosphere and can serve as a potential tool to predict ozone air quality. Shen et al. [173] estimated that in one half cycle of AMO (~35 years) from its cold to warm phase, the

summertime ozone levels in the US could increase by about 1–3 ppbv in the Northeast and 2–5 ppbv in the Great Plains. Yan et al. [223] also showed that AMO and ENSO indices could explain ~40% of the interannual variability of ozone concentrations in the US.

NAO and AO

Other climate oscillations such as the Arctic Oscillation (AO) and NAO have been found to influence tropospheric ozone at mid-high latitudes [28, 66]. In the positive AO phase, characterized by weaker sea level pressure in the polar region but higher sea level pressure at mid-latitudes, the weakened poleward transport from mid-latitudes to Arctic led to lower ozone (–1 DU) over the Arctic [165]. The variability of AO has shown to account for up to 50% of the observed ozone variability in the lower troposphere over North America in summer via changes in STE and intercontinental transport of ozone and its precursors [98]. The positive NAO phase intensifies the temperature gradient in the upper troposphere between mid-latitudes (~50° N) and high-latitudes (north of 60° N), and then affect the position of storm tracks and intensity. It is thus likely to strengthen STE [85] and influence surface ozone over Europe [143].

Synoptic Patterns

Monsoon

Monsoon is characterized by distinct seasonal transitions of prevailing wind and precipitation [33, 206]. The most energetic monsoon system is the Asian-Australian monsoon system spanning over the South and East Asia [33]. During winter, northerly wind prevails over South and East Asia, brings dry and cool weather conditions. The prevailing southwesterly with the onset of summer monsoon brings clean and moist ocean air to the continental southeast Asia, enhances cloud covers and precipitations. Convections are also active in the summer monsoon seasons.

Satellite and in situ observations have shown declines of tropospheric ozone in southeast Asia from May to August with the evolution of summer monsoon [155]. Significant ozone decreases over India could be attributed to transport pattern shifts, i.e., cleaner marine air input and stronger air uplift [156], and also lower ozone chemical production as a result of cloudy, cooler, and wetter weather conditions [135]. By quantifying the individual processes, Lu et al. [117] showed that the ozone chemical production decreased by 4.2 Tg over the Indian lower troposphere (from surface to 600 hPa) from May to August, and strong convection in August effectively uplifted 3.3 Tg ozone to above 600 hPa, together led to significant decreases in the Indian lower tropospheric ozone in the summer monsoon month. The uplifted

ozone in tropics can then be transported by the easterly jet in the upper troposphere and impact global tropospheric ozone distribution [96, 102, 106]. Similar ozone-monsoon responses but with different seasonal variations were also found for near-surface ozone in China [34, 62, 107, 118, 207, 240, 242].

Interannual ozone variability in monsoon regions shows strong correlations with the monsoon strength. Lu et al. [117] showed that ozone concentrations in the lower troposphere (from surface to 600 hPa) were 3.4 ppbv higher in weaker monsoon years than stronger years, mainly due to stronger ozone net chemical production. This negative correlation between ozone levels and monsoon strengths is also found at Pacific Ocean sites near the Asian continent [71]. Yang et al. [224], however, showed that stronger East Asian summer monsoons led to higher surface ozone concentrations over central and western China, mainly attributed to smaller ozone outflow to the East China Sea. Asian summer monsoon circulations are further modulated by climate variabilities such as ENSO [95] and AMO [114], and are projected to change in the warming future [157]. We thus expect these climate variabilities could also influence tropospheric ozone through change in monsoon on a longer timescale, which is still unknown due to the lack of long-term ozone observations.

Cyclone and Stagnation

The cold fronts associated with the mid-latitude cyclones can effectively lower air pollution [105, 197]. The frequency of ozone episodes in the northeastern US has showed a strong negative correlation with the cyclone frequency [105]. These cyclone activities are often related to the position of the polar jet wind. Combining observations and model simulations, Barnes and Fiore [12] found that the daily variability of US surface ozone was linked with the north-south latitudinal shift of the jet winds. Shen et al. [171] showed that the frequency of the jet wind traversing the Midwest and Northeast US acted as a good metric to diagnose the ozone variability in the northern US.

Surface ozone in Europe is strongly impacted by the strength and frequency of high-latitude blocks and subtropical ridges in summer [140]. A recent review from Dayan et al. [30] concluded that high summertime tropospheric ozone over the eastern Mediterranean could be attributed to frequent STE associated with tropopause folding activities [198], strong air subsidence at mid-troposphere [232], and the long-range transport of ozone-rich air masses from eastern continental Europe [154]. Myriokefalitakis et al. [128] suggested that the contribution of these dynamic processes (~90%) significantly outweighed that of local precursor emissions. High summertime ozone concentrations over the UK were often associated with anti-cyclonic conditions (degrading ventilation) and the easterly flows (transporting pollution from the continental Europe to the UK) [147].

Similar with front activities, stagnant conditions have been applied to diagnose air quality. Stagnations, which are usually characterized by slow wind speeds, no precipitation, and temperature inversion in the boundary layer, are unfavorable for ventilation and tend to build up high ozone air pollution [186]. High temperature events (heatwaves) could occur associated with stagnations under persistent high-pressure systems, leading to high ozone extremes [161, 172, 188]. Solberg et al. [179] summarized that during the 2003 Europe heatwave events, high ozone extremes were contributed by (1) extended air residence time in the stable boundary layer, (2) biomass burning due to drought and heat, (3) high biogenic isoprene emissions, and (4) reduced ozone dry deposition velocity. Sun et al. [188] showed that on average one stagnation day could increase the mean surface ozone concentration in the north-eastern US by about 4.7 ppbv.

Subtropical High

The semi-permanent subtropical high-pressure systems are mainly confined to oceans, but their intensifications in summer exert large influences on regional weather and air quality in regions such as the eastern US and eastern China [37, 43, 171, 239, 244]. Shen et al. [171] found that the influences of the Atlantic subtropical high (known as the Bermuda High) on ozone over the US depended on the location of its west boundary. The westward shift of the Bermuda High could increase ozone concentrations in regions under the high-pressure system, but decrease ozone along its west boundary by bringing clean and humid air from the ocean. Wang et al. [209] further showed that the location and strength of the Bermuda High explained 60–70% of the interannual variability of summertime ozone concentrations in the Houston–Galveston–Brazoria (HGB) metropolitan region. Focused on ozone air quality in China, Zhao and Wang [239] found that intensified West Pacific subtropical high enhanced southwesterly transport of moisture and clean air into South China, and therefore decreased ozone levels, but led to dry and sunny conditions over North China and thus increased ozone levels there.

Future Ozone Change Due to Climate Change

Previous sections have summarized three pathways of climatic influences on tropospheric ozone. In this section, we examine their combined effects in the context of future climate change. A review of future ozone projections driven by climate change was previously conducted by Jacob and Winner [82] and updated by Fiore et al. [46, 47] with more focus on the US. Here, we extend to more recent results (published after Jacob and Winner [82]), and include broader regional results.

Along with global warming driven by increasing greenhouse gas levels, there will be increases in the frequency,

duration, and intensity of regional hot extremes [25]. Hydrological cycle (water content, cloudiness, wet convections) will also respond to the warming. Global averaged specific humidity tends to increase due to more water vapor that can be accommodated in a warmer atmosphere, but relative humidity over land is expected to decline. There have also been some studies focusing on the future change of transport pattern (e.g., [36]). As discussed in the “[STE and Large-Scale Meridional Circulations](#)” section, increasing greenhouse gases will likely lead to accelerated stratospheric BDC and widening of the Hadley Circulation, which are expected to enhance STE in the future. Model projections show decreases of mid-latitude cyclone frequency due to poleward shift of polar jet stream over the eastern US in the twenty-first-century climate [103, 167, 227], and degradation of ventilation conditions with increasing stagnation days [70, 145].

Table 1 lists recent model projected future changes in surface or tropospheric ozone driven by climate change alone. The projections are from state-of-art chemical models with different frameworks (offline chemical transport model or on-line chemistry-climate model), model capability (dynamics, representation of natural emissions, and chemical mechanisms), model resolution, future greenhouse gas scenarios, time slice, and reported metrics. All these differences contribute to a wide range of projected ozone changes even for the same region [46, 47].

Despite different regional characteristics, most models predicted future climate change would lead to increases of surface ozone over polluted regions and decreases over remote land and oceans. Significant surface ozone enhancements were predicted in East Asia, Europe (in particular the southern Europe), and the northeastern US. Most models attributed surface ozone increases to warming-induced BVOC emission enhancements, faster chemistry kinetics, and also faster PAN decomposition. Only one result (Tai et al., 2013) listed in Table 1 included CO₂ inhibition on BVOC emissions. They showed that surface ozone enhancements would be reduced by 50% in major polluted regions when the CO₂ inhibition effect was included in the model. Over remote land and oceans, future surface ozone levels would generally decrease due to more water vapor and less PAN decomposition.

The different responses of surface ozone to future climate suggest “the most ozone polluted regions get worse while their neighbors get better” [162]. This is evident by more frequent occurrence of high ozone events (extremes) (e.g., [101, 104, 172, 208, 212]). For example, the 95th percentile of daily maximum 8-h average (MDA8) surface ozone in the US was projected to increase from 79 to 87 ppbv under the IPCC A2 scenario [146]. The increases of ozone extremes can be induced by a combined effect of higher ozone-temperature response in high NO_x regions [150], and more frequent and severe stagnations [70, 145] accompanied with persistent hot weather conditions [50, 172].

We highlight here the importance of increasing STE on future tropospheric ozone burden. Three projections ([11, 90]; and [122]) listed in Table 1 included stratosphere-resolved chemistry and dynamics in the models and thus better represent stratospheric influences on tropospheric ozone. All three models revealed significant enhancements of STE driven by stronger BDC, leading to increases of tropospheric ozone burden. Banerjee et al. [11] found that under the RCP8.5 scenario, climate change alone would indeed decrease net ozone chemical production (−109 Tg) due to higher water vapor content, but would then be compensated by increases of STE (+101 Tg), and result in a 13% increase of the tropospheric ozone burden. These results emphasized the need to better simulate STE in future ozone projections, however, many models (e.g., about half of the models in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) Phase 1; [99]) and most results listed in Table 1 still used prescribed stratospheric ozone as the lateral boundary or linearized stratospheric ozone schemes for future projections. Future studies are needed to understand to what extent the increasing STE influences future surface ozone air quality.

Feedback from Tropospheric Ozone Change to Climate

While tropospheric ozone is affected by climate change, its variations in turn influence climate through altering radiation and atmosphere-biosphere interactions. Using 17 different global climate-chemistry models with varying radiation schemes, Stevenson et al. [184] estimated the tropospheric ozone radiative forcing of 410 mW m^{−2} from the pre-industrial era (1750) to 2010. Compared to CO₂ and methane, the shorter lifetime of tropospheric ozone leads to heterogeneous spatial distributions of its abundance and resulting radiative forcing. The highest tropospheric ozone radiative forcing values are found over the northern mid-latitudes where the sources of ozone precursors are large, and over cloudless subtropical regions such as the Sahara Desert where vertical temperature differences are high [184].

The heterogeneous distribution of ozone radiative forcing may alter atmospheric general circulation. High tropospheric ozone and black carbon levels at the northern mid-latitudes intensify the meridional temperature gradient in the UTLS, and partly drive the observed expansion of the Northern Hemisphere tropics [2]. As discussed in the “[STE and Large-Scale Meridional Circulations](#)” section, widening of the tropics (also the Hadley Circulation) may further increase tropospheric ozone, providing a potential positive feedback [120]. Exclusion of ozone radiative feedback in CCMs would also cause models to predict stronger weakening of the Walker circulation and more ENSO extremes in the future [133]. In

Table 1 Recent (since 2009) projections of future tropospheric ozone driven by climate change (abbreviation for model type: CM = climate model, RCM = regional CM, CTM = chemical transport model, RCTM = regional CTM, GCM = general circulation model, CCM = climate chemistry model; abbreviation for scenario: IPCC A1B/A2 scenarios [129], RCP 2.6/4.5/6.0/8.5; Representative Concentration Pathways (RCP) scenarios with radiative forcing of 2.6/4.5/6.0/8.5 W m⁻² by 2100 [202]; abbreviation for metrics: MDA8 = daily maximum 8-h average, JJA = June–July–August, DJF = December–January–February; abbreviation for regions: NE = northeast, SE = southeast, IMW = Intermountain West, NA = North America, EU = Europe, EA = East Asia, SA = South Asia; note that domains can be different among studies)

Reference	Type (resolution)	Region	Scenario	Time horizon	Metric	Ozone change (uncertainty if provided)	Important contribution factors (with +/- signs denoting directions of global ozone change)
Kawase et al. [90] #	CCM (2.8°×2.8°)	Globe	RCP 8.5	2100 vs. 2005	Annual mean tropospheric ozone column (DU)	+ 5.5	1) Increase in STE (+) 2) Higher water vapor concentrations (-)
Doherty et al. [35]	3 CCMs (5°×5° to 3.75°×2.5°)	Globe	IPCC A2	2095–2099 vs. 2001–2005	Annual surface average ozone (ppbv or %)	NA: -2.2% (-1.4 to -3.4%) ^a EU: -1.0% (-2.0 to +0.4%) EA: -1.6% (-2.9 to +0.2%) Polluted region: +6 ppbv	Globe: 1) Decrease in PAN decomposition (-) 2) Higher water vapor concentrations (-) Polluted regions: 1) Increase in PAN decomposition (+) 2) Increasing BVOC emissions (+) 3) Higher water vapor concentrations (-)
Tai et al. [191]*	GCM-CTM (5°×4°)	Globe	IPCC A1B	2050 vs. 2000	Surface summertime MDA8 ozone (ppbv)	NA, EU, EA: maximum of +6 (NOT include CO ₂ inhibition) NA, EU, EA: maximum of +3 (include CO ₂ inhibition)	1) Increase in PAN decomposition (+) 2) Increasing BVOC emissions (+)
Banerjee et al. [11] #	CCM (3.75°×2.5°)	Globe	RCP 8.5	2100 vs. 2000	Annual mean tropospheric ozone burden (Tg or %)	+43Tg or 13.1%	1) Increase in STE (+) 2) Higher water vapor concentrations (-)
Schnell et al. [162]	4 CCMs (2°×2° to 3.75°×2.5°)	Globe	RCP 8.5	2100 vs. 2000	Surface summertime MDA8 ozone (ppbv)	West NA: -0.2 (-2.1 to +5.0) ^a East NA, +1.8 (-2.2 to +7.3) South EU: +2.0 (-1.3 to +9.3) North EU: -0.9 (-3.9 to +2.0) South EA: -2.8 (-4.7 to -0.8) North EA: -0.5 (-2.5 to +3.1) +112 Tg or 28%	1) Increasing BVOCs emissions (+) 2) Faster kinetics (+) 3) More stagnations (+) 4) Higher water vapor concentrations (-)
Meul et al. [122] #	CCM (2.8°×2.8°)	Globe	RCP8.5	2100 vs. 2000	Tropospheric ozone burden (Tg or %)		1) Increase in STE (+)

Table 1 (continued)

Reference	Type (resolution)	Region	Scenario	Time horizon	Metric	Ozone change (uncertainty if provided)	Important contribution factors (with +/- signs denoting directions of global ozone change)
Hedegaard et al. [64]	GCM-RCTM (N/A)	NH	RCP 4.5	2090–2099 vs. 1990–1999	Annual surface ozone (%)	Arctic: +5 to +10 ^b EU, US, and SE Asia: +5 to +20 Remote: -10 to -5	1) Higher ozone import to Arctic (+) 2) Less ozone dry deposition in the Arctic (+) 3) Increasing BVOC emissions (+) 4) Higher water vapor concentration (-)
Andersson and Engardt [5]	RCM-CTM (0.44°×0.44°)	Europe	IPCC A2	2071–2100 vs. 1961–1990	April–September surface daily maximum ozone (ppbv)	-3 to +25 ^b	1) Decreasing dry deposition (+) 2) Increasing BVOCs emission (+)
Katragkou et al. [88]	RCM-RCTM (50×50 km)	Europe	A1B	2091–2100 vs. 1991–2000	Median JJA surface ozone (ppbv)	+3.9 to +6.2 ^b	1) Decreasing cloudiness (+) 2) More stagnant condition (+) 3) Increasing BVOC emission (+)
Langner et al. [101]	5 GCM-RCM-RCTM/RC-CM (150×150 to 50×50 km)	Europe	A1B	2040–2049 vs. 2000–2009	Maximum positive changes in April–September surface mean ozone (ppbv)	+2.7 (+1.2 to +3.0) ^a	1) Increasing BVOC emission (+)
Colette et al. [23]	GCM-CTM (3.75°×2.5°)	Europe	RCP8.5	2045–2054 vs. 1996–2005	JJA surface MDA8 ozone (μg m ⁻³)	Below +1	1) Increasing BVOC emissions (+)
Colette et al. [24]	11 all-type models (5°×5° to 0.44°×0.44°)	Europe	Multi-scenario	2070–2100 vs. 2000s	JJA surface ozone (ppbv)	EU: +1.25 (+0.99 to +1.5) ^c Alps: +5 JJA: +0.36 (-0.11 to +0.83) ^a DJF: +0.05 (-0.21 to +0.26)	1) Increasing BVOC emissions (+)
Watson et al. [211]	4 GCM-RCTMs (50×50 km)	Europe	RCP 4.5	2050 vs. 2006	Surface mean ozone (ppbv)	No dominant drivers are concluded	
Avisé et al. [8]	RCM-CTM (36×36 km)	US	IPCC A2	2045–2054 vs. 1990–1999	July surface MDA8 ozone (ppbv)	NE US: +1 to +4 ^b SE US: -6 to -1	1) NE US: increasing temperature (+) 2) SE US: increasing precipitation decrease organic nitrates (-)
Weaver et al. [212]	12 all-type models (4–36 km; 0.44°×0.44° to 5°×4°)	US	Multi-scenario	End of twenty-first century vs. present	JJA surface MDA8 ozone (ppbv)	NE US: 0 to +4 ^b IMW: -2 to +3 SE US: -6 to +5	1) Increasing BVOCs emissions (+) 2) Increasing solar radiation and temperature (+)
Lam et al. [97]	GCM/RCM-RCTM (12×12 km)	US	A1B	2050 vs. 2000	Annual surface MDA8 ozone (ppbv)	+2.0 to +2.5 on average	1) Increasing BVOCs emissions (+)

Table 1 (continued)

Reference	Type (resolution)	Region	Scenario	Time horizon	Metric	Ozone change (uncertainty if provided)	Important contribution factors (with +/- signs denoting directions of global ozone change)
Kelly et al. [91]	RCM-RCTM (45 × 45 km)	US	A2	2041–2050 vs. 1997–2006	JJA surface MDA8 ozone (ppbv)	+9 to +10 in urban region	1) Increasing temperature and solar radiation (+) 1) NE US: decrease in cyclone frequency (+) 2) IMW US: higher water vapor concentrations (-)
Clifton et al. [22]	CCM (2°×2°)	US	RCP8.5	2091–2100 vs. 2006–2015	JJA surface ozone (ppbv)	NE US: +3 IMW US: -4 to -1 ^b Maximum of +10	1) Increasing solar radiation and decreasing cloudiness (+) 2) Increasing BVOC emission (+) 1) Decrease in cyclone frequency (+)
Pfister et al. [146]	GCM-RCCM (36 × 36 km)	US	A2	2046–2058 vs. 1996–2008	JJA surface MDA8 ozone (ppbv)	Maximum of +10	1) Increasing solar radiation and decreasing cloudiness (+) 2) Increasing BVOC emission (+)
Rieder et al. [151]	CCM (48 × 48 km)	Eastern US	RCP 4.5	2091–2100 vs. 2005	JJA surface MDA8 ozone (ppbv)	+1 to +2 on average, maximum of +4	1) Decrease in cyclone frequency (+)
Gonzalez-Abraham et al. [54]	GCM-RCTM (36 × 36 km)	US	A1B	2045–2054 vs. 1995–2004	JJA surface MDA8 ozone (ppbv)	NW US: -1.0 Other regions in US: +0.4 to 7.2	NWUS: decreasing in solar radiation (-) Others: 1) Increasing solar radiation (+) 2) Increasing temperature (+)
Val martin et al. [201]	CCM (2.5°×1.9°)	US	RCP 8.5	2050 vs. 2000	Annual surface MDA8 ozone (ppbv)	US: +2 Eastern US: maximum of +5	1) Increasing BVOCs emissions (+) 2) Decreasing dry deposition velocity (+) 3) Decreasing precipitation (+)
Nolte et al. [132]	GCM-RCM-CTM (1.25°×0.9°)	US	RCP 8.5	2030 vs. 2000	JJA surface MDA8 ozone (ppbv)	+0.2 to +2.9 ^b	1) Increasing BVOC emissions (+) 2) Decreasing BVOC emissions (+)
Wang et al. [208]	GCM-CTM (5°×4°)	China	A1B	2050 vs. 2000	Annual surface mean ozone (ppbv)	Eastern China: +0.5 to +3 ^b Western China: -2 to -0.1	1) Eastern China: increasing BVOCs emission (+) 2) Western China: higher water vapor concentrations (-)

*The model considered CO₂ inhibition for biogenic isoprene emission as discussed in the “BVOC Emissions” section

#The study considered full stratosphere dynamics and chemistry and discussed the impact on tropospheric ozone change

^aNumbers are ranges among individual models

^bNumbers are ranges among different regions or model grids

^cNumbers are 95% confidence interval

addition, tropospheric ozone can influence the radiative forcing of other chemical tracers such as methane and NO_x by changing their lifetimes [44, 47].

Tropospheric ozone also affects climate indirectly through its impacts on vegetation and carbon uptake [113, 177]. Stomatal uptake of ozone damages plant cells and impedes plant photosynthesis, leading to reductions of plant primary productivity [1, 39, 231]. Sitch et al. [177] estimated that under the IPCC A2 scenario, increasing tropospheric ozone in 2100 would decrease the global gross primary productivity by up to $30 \text{ Pg C year}^{-1}$ compared to the 1990 condition, exerting indirect radiative forcing of 1.09 W m^{-2} . The declined vegetation would decrease the amount of BVOC emissions, and therefore limit ozone production, but it would also suppress ozone dry deposition. Such interactions between climate, atmospheric chemistry, and the biosphere are still poorly understood and are generally not considered in current studies.

Conclusion

Variations and future changes of tropospheric ozone are strongly tied to meteorology and climate (Fig. 1). Meteorology influences the biogenic activities of vegetation and microbes in the ecosystem and hence their emissions of ozone precursors. These climate-sensitive natural emissions mainly include soil NO_x , lightning NO_x , BVOCs, wildfires, and wetland methane emissions. Meteorology also determines the nature of atmosphere where photochemistry relies on, and therefore influences tropospheric ozone through altering the kinetics, and partitioning and deposition of chemicals. Changes in atmospheric circulation on different spatiotemporal scales influence the transport of ozone and its precursors. In particular, robust signals of ozone response have been found to large-scale circulations (e.g., BDC) and STE, large-scale climate patterns (e.g., ENSO, AMO), and synoptic patterns (e.g., monsoon, cyclones). All these connections together determine the high sensitivity of tropospheric ozone levels to climate.

Projections of future ozone changes driven by climate change largely reflect the dominant role of increasing temperature and water vapor in the atmosphere. These suggest increasing surface ozone in the polluted regions such as eastern US, southern Europe, and the south and east Asia, most likely due to increasing biogenic isoprene emissions, increasing solar radiation with less cloudiness, decreasing ozone dry deposition, increasing PAN decomposition, and higher frequency of stagnations and heat waves. Additional emission control measures are thus required over such regions to meet the ozone air quality standards in the future. In remote regions and ocean, surface ozone levels are projected to decrease due to stronger chemical loss with higher water vapor and also less PAN decomposition. The change of tropospheric ozone

burden can be affected by the competing roles of increasing water vapor (which decreases tropospheric ozone) and increasing STE due to stronger BDC (which increases tropospheric ozone).

Considerable limits still exist in the current understanding of the biogenic, chemical, and dynamic linkages between ozone and climate, which challenge our confidence in the model projections of future ozone change. Previous reviews have raised some major recommendations, e.g., improving the capability of climate models to present local processes, constraining uncertainties in atmospheric chemistry mechanisms (in particular the uncertain yield and fate of isoprene nitrates), and using ensemble model runs for future projections [46, 47, 82]. Here, we prioritize two important issues for further research and model development.

1. Uncertainties in biogenic activities and their responses to changing environment. The ecosystem serves as a hub to connect tropospheric ozone and climate, yet their linkages need to be better understood. Models may not adequately present many of these biogenic activities, for example, the inhabitation of BVOC emissions with rising CO_2 levels [191], biogenic isoprene emissions in rapid transition of weather conditions (e.g., [235]), and ozone damage on vegetation (further influence emission of BVOCs and uptake of ozone and carbon). Many models also do not consider the climate-induced terrestrial change (e.g., evolution of plant types and land cover), which has important implications for the ozone variation as many of the terrestrial responses are dependent on plant types [26]. Improved scientific knowledge as well as the development of fully coupled earth system models is in need to better quantify such interactions.

2. The role of future stratospheric circulation and STE on tropospheric ozone. As discussed in the “[Future Ozone Change Due to Climate Change](#)” section, models that predict stronger stratospheric BDC in the future show notable increases in tropospheric ozone burden driven by changes of STE, while models with no or inadequately stratosphere dynamics predicted tropospheric ozone decreases. Coupling with future stratosphere ozone recovery [32, 180], stronger STE may become a key factor modulating future tropospheric ozone and even surface ozone. Representing these dynamic ozone responses requires models to include stratosphere-resolved dynamics and chemistry.

Finally, we also briefly review the feedback of tropospheric ozone to climate change through exerting RF and interactions with biosphere. The heterogeneous spatial distribution of tropospheric ozone exerts notable influences on the global and regional scale atmospheric circulations such as the Hadley Circulation and the Walker Circulation. The increasing surface ozone also impedes the carbon uptake in ecosystem and therefore indirectly influence climate. A comprehensive view of the interactions between tropospheric ozone, ecosystem, and radiation remains to be quantified in future studies.

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Compliance with Ethical Standards

Conflict of Interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

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