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Tropospheric ozone and its natural precursors impacted by climatic changes in emission and dynamics

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Climate change plus ozone pollution is a lethal combination that adversely affects human health, agriculture productivity, ecosystems, and the world economy. Currently, there is a growing concern that climate change may aggravate ozone pollution even after reducing anthropogenic precursor source activities. Part of it may be due to temperature-enhanced natural precursor emissions of ozone, increased stratospheric–tropospheric exchange of ozone, more frequent periods with meteorologically stagnant conditions favouring pollution build-up, changes in wetland methane emissions, and enhanced concentrations of atmospheric carbon dioxide. The quantification of climate change and ozone levels involves complex chemical, radiative, biogenic, and transport processes and feedback that are difficult to fully comprehend. Further, the fact that ozone is a short-lived climate forcer and a potent greenhouse gas provides a feedback loop and makes climate-ozone links even more multifaceted. This review examines the up-to-date understanding of the processes regulating tropospheric ozone from regional to global scales and the associated climate connections. We explore the recent findings in ozone precursor emissions from natural sources, ozone formation chemistry, its transport on hemispheric scales, future plausible ozone concentrations in different shared socioeconomic pathways, and changes in the radiative forcing of ozone. The review demonstrates the challenges and limitations associated with climate-ozone linkages and their incorporation in models, which are due to uncertainties in magnitude and signs of projected precursor emissions in response to future climate change and also due to the difference in models.

KEYWORDS

tropospheric ozone, ozone precursors, climate change, long-range transport, BVOCs, ENSO, LNO$_x$

Introduction

Climate and air quality are inseparably linked. Air pollutants like ozone and particulate matter interact with long-wave and short-wave radiations and disturb the energy balance of the Earth, contributing to climate change. Climate change in turn influences these pollutants by altering the frequency, severity, and duration of heat waves, air stagnation events, precipitation, anticyclonic conditions, air pollution meteorology (ventilation and dilution), and other removal processes (Fiore et al., 2015; Doherty et al., 2017). Climate change can induce magnifying responses in atmospheric chemistry, emissions from anthropogenic and natural sources of these pollutants, and also impacts their long-range transport. This paper focuses on how climate change impacts the emissions and dynamics of tropospheric O_3 and its natural precursors.

Ozone is one of the most significant trace gases in the atmosphere. It is a strong oxidant, a short-lived climate forcer (SLCF), and an important greenhouse gas (IPCC Pörtner et al., 2022). In the stratosphere, ozone plays an important role in protecting us from harmful ultraviolet radiation. However, in the troposphere, ozone is a pollutant that is detrimental to human health, crops, and ecosystems (Monks et al., 2015).

Both short-term and long-term exposure to ozone is associated with increased mortality due to respiratory and cardiovascular diseases. Ozone-related deaths are assessed to be 5–20% of all those associated with air pollution (Anenberg et al., 2009; Silva et al., 2013; Lin et al., 2018). Moreover, tropospheric ozone (O_3) is a well-known phytotoxin that has an adverse impact on natural vegetation and crops (Feng et al., 2020). Different crop yield metrics have been used extensively to evaluate global yield losses due to present-day O_3 concentrations, which are estimated to be 2–16% for the four major staple crops (wheat, rice, maize, and soya bean; (Ainsworth, 2017). Associated economic losses due to ozone pollution are estimated to be 32 billion in China alone (Xu, 2021).

Ozone in the troposphere is formed by photochemical reactions involving sunlight and ozone precursor gases including nitrogen oxides (NO_x) and methane, carbon monoxide (CO), and non-methane volatile organic compounds (NMVOC; (Kinney, 2018), (Jacob, 2000)). These precursors originate from natural sources including wetland methane emissions, wildfires, biogenic hydrocarbon emissions, lightning NO_x, and NO_x emitted from soils, but also anthropogenic activities like fossil fuel combustion in power plants, industrial activities, transportation, or crop burning. The Sustainable Development Goals are pressing nations to impose emission reductions and curb fossil fuel emissions. In such a scenario, the emission of natural ozone precursors will gain increasing importance in regulating future ozone concentrations (Cai et al., 2021).

The amount of O_3 produced is dependent on precursor concentrations and can be categorized into photochemical regimes such as NO_x-limited or VOC-limited. In NO_x-limited regimes, ozone increases with increasing NO_x and exhibits only slight sensitivity to VOC whereas in a VOC-limited regime reduction in VOC emissions can result in decreases in O_3 levels, but NO_x reduction has a weak effect or may even increase ozone levels (Sillman and He, 2002). O_3 formation in polluted urban areas is usually reported as VOC-limited, while that in rural areas falls in NO_x-limited. Besides these two regimes, a third regime is gaining importance in which the reactive uptake of hydroperoxyl radicals onto aerosol particles suppresses the surface ozone concentrations and inhibits its formation (Ivatt et al., 2022). Peroxyacetyl nitrate (PAN) is a significant reservoir of NO_x, with a lifetime of almost a month in the free troposphere, and its transport at an intercontinental distance allows O_3 production to occur far away from its source (Jiang et al., 2016).

Changes in meteorology and climate impact O_3 levels by affecting natural emissions of its precursors like NO_x from the soil, lightning, and biogenic volatile organic compounds (BVOCs). The rise in ambient temperature can accelerate the emission rate of BVOCs and soil NO_x (Sha et al., 2021). The dominant mechanism of ozone formation in the upper troposphere is lightning-induced nitrogen oxides (NO_x) which are affected by increasing temperatures (Verma et al., 2021). The growth in wetland methane emissions can be a major driver for increases in background ozone in the coming years. Wetland methane emissions are also sensitive to elevated temperatures and CO_2 concentrations (Shindell et al., 2004).

In the troposphere, the lifetime of ozone ranges from a few hours in polluted urban areas to up to a few months in the upper troposphere. The global mean lifetime of ozone for the present day is estimated to be around 25 days, making its transfer possible not only on regional but also over intercontinental scales (IPCC Pörtner et al., 2022). Long-range and regional transport of ozone can also upsurge local O_3 levels (Yang et al., 2022). Variations in stratosphere troposphere exchange (STE) and large-scale climate patterns such as the El Niño–Southern Oscillation (ENSO), North Atlantic Oscillation (NAO), and Arctic Oscillation (AO) have substantial impacts on present-day ozone distribution and ozone projections in emerging years (Albers et al., 2022; Yang et al., 2022). Planetary boundary layer (PBL) growth plays a critical role in the development of elevated O_3 episodes. especially when there is no pollutant transport from a long distance. Studies report the downward transport of ozone from high ozone concentration layers with boundary layer development in the morning and afternoon and this has been found to aggravate near-surface ozone pollution (Dey, 2018; He et al., 2021).

High ozone concentrations are typically associated with hot, dry, stagnant conditions as the formation and persistence of O_3 is favoured (Zhan and Xie, 2022). Studies have demonstrated ozone

extremes frequently appear during drought besides heat waves and at times this coincidence is due to specific synoptic patterns. Heightened periods of air mass stagnation and rising temperatures in the future would lead to increased O_3 concentrations (Li et al., 2020). Increased water vapour and precipitation in a changing climate are expected to decrease ozone concentrations (Sekiya and Sudo, 2012).

To combat ozone pollution efforts have been made to cut anthropogenic emissions of ozone precursors NO_x and VOCs. However, the reductions at local and regional levels of the emissions of pollutants do not always imply a decrease in the concentrations of other pollutants as the processes linking emissions and air quality are interconnected and quite complex (Li et al., 2013). It has been reported that a 40% reduction in particulate matter concentrations was partially responsible for a 1–3 ppb increase in ozone levels in eastern China (Zhang et al., 2019).

Ozone interacts with long-wave and short-wave radiation and perturbs the earth's radiative balance contributing to climate change. Climate change in turn affects natural sources, chemistry, and transport of ozone and its precursors. Short-term climate variability alters the long-range transport pathways that channel ozone plumes on a regional and intercontinental scale (Mann, 2021).
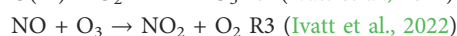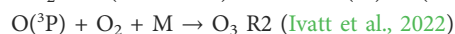
Stringent air quality improvement measures in various countries have resulted in the decrease of anthropogenic precursor emissions in many regions. In such a scenario role of biogenic precursors in regulating ozone levels may become more significant. Thus, it is important to examine the impact of climate change on natural emission sources of ozone along with the effect on photochemistry, transport, and changes in synoptic-scale meteorology. This has important consequences for future ozone projection in the context of climate change.
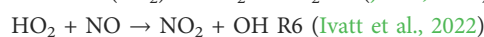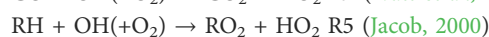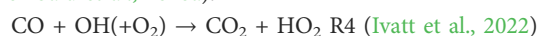
Ozone chemistry in the troposphere

Ozone is found both in the stratosphere and troposphere. Around 90% of ozone is found in the stratosphere mainly in the ozone layer (~between 8 and 15 km above the ground). There is a natural transport of ozone from the stratosphere to the troposphere that was earlier considered the primary source of tropospheric ozone (Archibald et al., 2020a). This transport contributes to background levels of ground-level ozone in the troposphere but ozone is also produced photochemically throughout the troposphere. Present-day estimates state that photochemistry is the main source of ozone in the troposphere, exceeding the flux from the stratosphere by factors of 7–15 (Cooper et al., 2014).

It is well known that the photolysis of NO_2 is the fundamental reaction leading to the formation of tropospheric

ozone (R1, R2). Once formed ozone reacts with NO present in the atmosphere to regenerate NO_2 (R3).

$NO_2 + hv \ (\lambda < 424 \ nm) \rightarrow NO + O \ (^3P)$ R1 (Ivatt et al., 2022)
$O(^3P) + O_2 + M \rightarrow O_3$ R2 (Ivatt et al., 2022)
$NO + O_3 \rightarrow NO_2 + O_2$ R3 (Ivatt et al., 2022)

The net ozone production occurs due to the presence of VOCs viz CO, CH_4, and other hydrocarbons, which are oxidized by OH radicals to produce hydroperoxyl and organic peroxy radicals (R4, R5). These radicals are the main intermediates in tropospheric ozone production as they provide an additional pathway to convert NO to NO_2 without destroying ozone (Archibald et al., 2020a).

$CO + OH(+O_2) \rightarrow CO_2 + HO_2$ R4 (Ivatt et al., 2022)
$RH + OH(+O_2) \rightarrow RO_2 + HO_2$ R5 (Jacob, 2000)
$HO_2 + NO \rightarrow NO_2 + OH$ R6 (Ivatt et al., 2022)

The chain termination can occur either through the self-reaction of peroxyl radicals to form longer-lived species (such as peroxides, R7) or from the reaction of hydroxyl radicals with nitrogen dioxide (NO_2) to form nitric acid (R8).
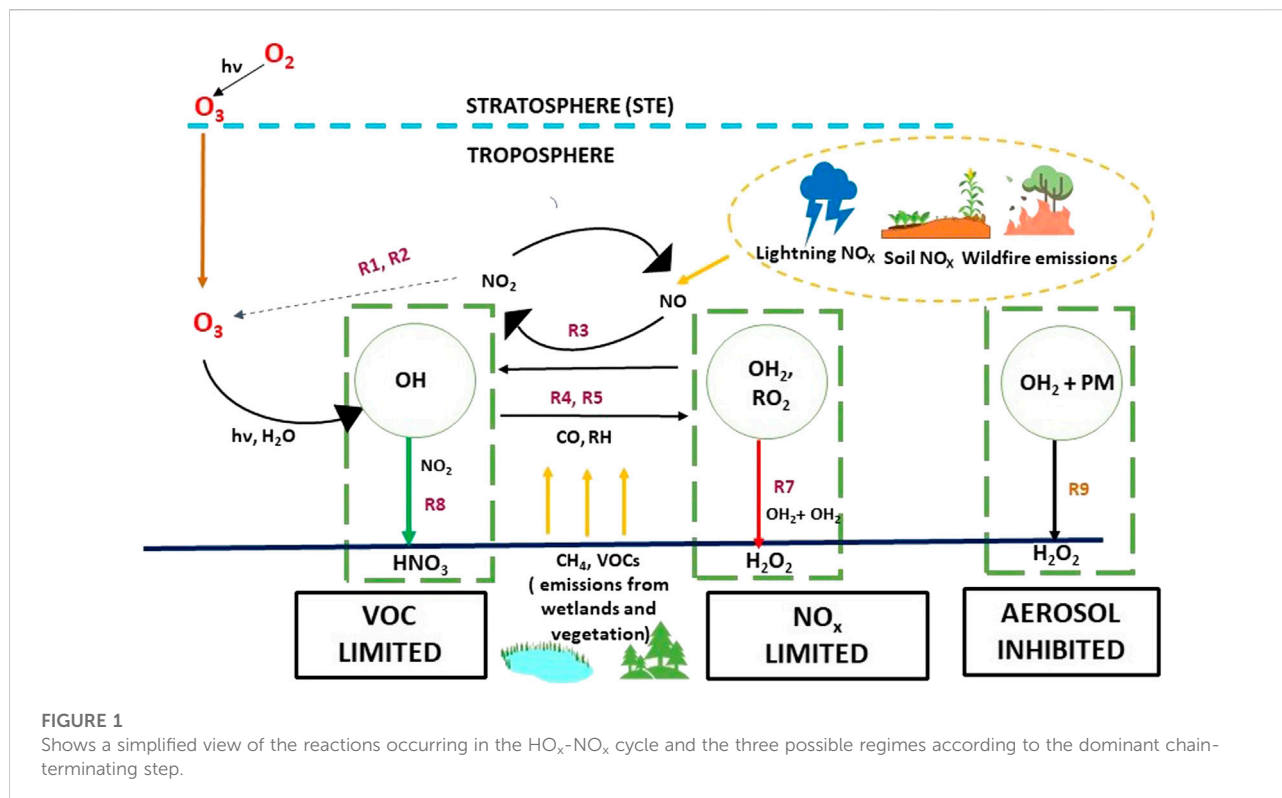
$OH_2 + OH_2 + M \rightarrow H_2O_2 + O_2$ R7 (Jacob, 2000)
$OH + NO_2 + M \rightarrow HNO_3 + M$ R8 (Jacob, 2000)
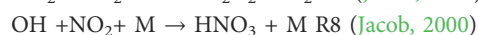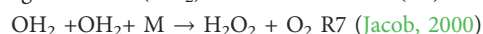
The controlling chain termination step differentiates regions into either "NO_x limited" (peroxyl-radical self-reactions dominate) or 'VOC limited' (hydroxyl radical + nitrogen dioxide reaction dominates; (Thornton et al., 2002)). For reducing O_3 pollution, the dominant termination step is frequently used to identify whether reductions in the emissions of NO_x or VOCs should be preferentially employed. If peroxyl-radical self-reactions dominate (known as the 'NO_x limited' regime), reductions in NO_x emissions are most beneficial, whereas if the NO_2+OH reaction dominates (known as the "VOC limited" regime) reductions in organic emissions are most effective (Figure 1).

Recent studies using the chemical transport model show that a third aerosol-inhibited regime exists especially in large regions of India and China where the presence of aerosols inhibits ozone formation on short photochemical timescales (Archibald et al., 2020a). In this regime reactive uptake of hydroperoxyl radicals onto aerosol particles dominates the chain termination step and the presence of aerosol inhibits ozone formation (Xu et al., 2012; Ivatt et al., 2022). Xu et al. reported that the HO_2 heterogeneous reaction decreased O_3 to 33 ppb in Beijing during summer.

$HO_2 + PM \rightarrow 0.5H_2O_2$ R9 (Xu et al., 2012)
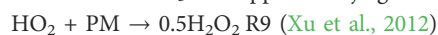
The HO_2 reaction with aerosols would terminate the HO_x cycle limiting ozone formation via NO_x. The uptake coefficient of hydroperoxyl radical on aerosol is affected by relative humidity and aerosol composition. The overall impact of change in aerosol concentration on ozone levels is complex as it depends on several other processes besides photochemical reactions. Li et al. report a decrease in $PM_{2.5}$ was a chief factor driving the increase in ozone in China due to the role of $PM_{2.5}$ as a scavenger of hydroperoxyl (HO_2) radical (Li et al., 2020).

FIGURE 1
Shows a simplified view of the reactions occurring in the HO_x-NO_x cycle and the three possible regimes according to the dominant chain-terminating step.

Peroxyacetyl Nitrate Chemistry

PAN is the principal reservoir species for NO_x in the troposphere. The OH reaction with acetaldehyde is the most important way to form PAN globally (Archibald et al., 2020a). The peroxyacetyl radical, $CH_3C(O)OO$, reacts with NO_2 to form PAN. PAN is a thermally unstable compound; its lifetime depends on the temperature. At higher temperatures, the PAN lifetime becomes shorter and the decomposition of PAN into peroxy acetyl radicals ($CH_3C(O)O_2$) and NO_2 is more rapid.

Therefore, at high temperatures, PAN becomes a source of NO_x rather than a reservoir of NO_x. This may then lead to an increase in the rate of ozone production (Archibald et al., 2020b; Zhang et al., 2021). Kotchenruther et al., and Liu et al. have shown that the thermal decomposition of PAN plays a crucial role in determining ozone concentrations (Kotchenruther et al., 2001; Liu et al., 2021). The lifetime of PAN against thermal decomposition is about 2 h at a typical NO_2/NO ratio of ~7 and ambient temperature of 25°C but it rises rapidly with the decrease of temperature, approximately five times every 10°C, extending it to over a month in the mid-troposphere (Liu et al., 2021). At the temperature of the upper troposphere, PAN acts as the main reservoir for short-lived NO_x, and thus facilitates the transport and release of NO_x to remote regions (Sun et al., 2021), with important implications for global O_3 distribution.

Effect of climate change on ozone precursor emissions by natural systems

Natural emissions of ozone precursors from the surface significantly impact ozone levels (Archibald et al., 2020a). Climate-related changes in lightning NO_x emissions, soil NO_x, wetland methane emissions, biogenic volatile organic compound emissions, and wildfire emissions content are significant drivers of future tropospheric ozone changes, affecting its chemical production and loss processes. In this section, we summarise the recent studies on the effect of climate change on these ozone precursor emissions by natural systems.

Soil NO_x

NO_x is formed as a by-product of nitrification and denitrification, two microbial processes that occur in many natural and agricultural ecosystems. Nitrogen oxides emitted from soils contribute 10–15% of global NO_x emissions (Weng et al., 2020). Weng et al. reported global total above-canopy soil NO_x emissions to be 3.3–10 Tg N/yr in bottom-up studies and at 7.9–16.8 Tg N/yr for satellite-based top-down studies (Weng et al., 2020). Lu et al. suggest that moisture, soil texture, temperature, nutrient availability, fertilizer application time

and method, type of vegetation, and ecosystem affect NO_x emissions from soils (Lu et al., 2019; Lu et al., 2021). With control of anthropogenic emissions of NO_x in the future, SNO_x may play an important role in increasing ozone levels, particularly in a changing climate. Geddes et al. reported SNO_x emissions to make up ~25% of summertime NO_x emissions in the United States in the "low" anthropogenic NO_x emission scenario (Geddes et al., 2022).

Romer et al. reported the ozone enhancements on hotter days and evaluated that SNO_x emissions are accountable for almost half of the 2.3 ppb/°C increase in O_3 production rates with ambient temperatures in the southeast United States (Romer et al., 2018). This can be attributed to temperature-enhanced SNO_x emissions most probably by soil microbes. Enzymatic activity increases with rising temperatures and a 40% increase in nitrogen fixation are expected in comparison to 2000 (IPCC Pörtner et al., 2022).

In ecosystems with broadly variable temperatures, NO_x emissions may be affected by temperature mainly during periods of optimal soil moisture and high inorganic nitrogen concentrations. SNO_x can increase by 38% on average as the soil temperature increase from 30–35°C to 35–40°C (Sha et al., 2021). SNO_x emission perturbations lead to changes in O_3 that are more regional.

Lu et al. reported presence of soil NO_x emissions in China significantly reduced the sensitivity of ozone to anthropogenic emissions. The study reported that the control of all domestic anthropogenic air pollutants to achieve maximum improvement in air quality decreased by 30% due to the presence of SNO_x (Lu et al., 2021). Sha et al. estimated that 40.1% of total NO_x emissions in California were from soils, and SNO_x significantly increased the monthly mean NO_2 columns by 34.7% and surface NO_2 concentrations by 176.5%, which led to an additional 23.0% of surface O_3 concentration in California (Sha et al., 2021).

SNO_x emissions are also likely to be controlled by changes in precipitation patterns and trans-evaporative fluxes (Lu et al., 2021; IPCC Pörtner et al., 2022). These SNO_x emissions are usually estimated through non-linear parameterization which is sensitive to the horizontal resolutions of inputted meteorological and ancillary data. Weng et al. in a sensitivity test study estimated that a 1°C increase in temperature would lead to a 5.2% increase in the calculated global total SNO_x emission, a 10% increase in soil moisture would lead to a 15.8% decrease in emission, and a 10% increase in leaf a would lead to 1% decrease in emission (Weng et al., 2020).

It is evident that SNO_x emissions will increasingly contribute to high O_3 episodes possibly changing with agricultural phenology, land management, precipitation or soil wetness, and temperature but suitable representations of SNO_x and how it escapes from the canopy are still lacking in models to provide quantitative evaluations of climate-driven changes in SNO_x (Lu et al., 2021).

Lightning NO_x

Lightning contributes ~10% of the total NO_x emissions and is the key source of NO_x in the middle to upper troposphere. Globally lightning generates NO_x equivalent to 2–8 Tg N/yr (Kang et al., 2020). During lightning, high temperatures result in the dissociation of O_2 to form O atoms. This initiates the production of NO through R10-R12.

$O_2 \rightarrow O + O$ R10 (Chameides, 1986)
$O + N_2 \rightarrow NO + N$ R11 (Chameides, 1986)
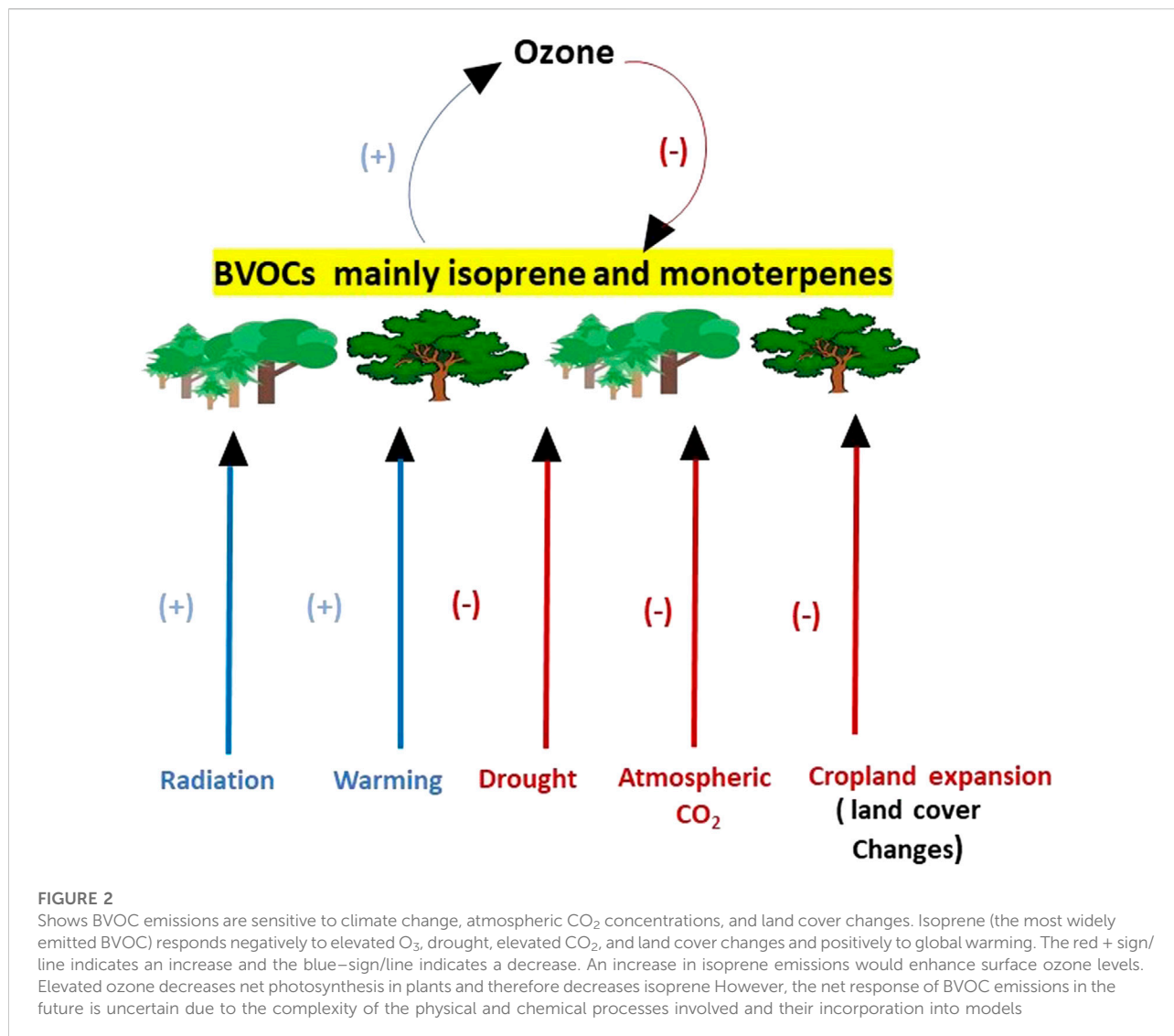$N + O_2 \rightarrow NO + O$ R12 (Chameides, 1986)

During the interconversion of NO and NO_2, ozone is formed in presence of hydroxy and peroxy radicals as discussed in the earlier section. Ozone can undergo photodissociation and react with water vapour to generate hydroxyl radicals (Bucsela et al., 2019). The OH radical formed is a strong oxidant that can decrease the lifetime of CH_4 which can reduce ozone levels. It is estimated that LNO_x is responsible for 35–45% of global free-tropospheric ozone production (Zhang et al., 2022a).

The impact of lightning is strong in the middle and upper part of the troposphere as in this region NO_x has a lifetime of 5–10 times longer than the ~1-day lifetime in the lower troposphere. Ozone production ensues at rates of up to 10 ppbv per day in the lightning-enhanced convective outflow plumes of ozone precursors. In the upper troposphere, the lightning source dominates, and estimates report LNO_x accounts for 60–70% of free-tropospheric NO_x (Bucsela et al., 2010). Ozone is the third most significant greenhouse gas, and ozone enhancements near the tropopause have the greatest effect on its radiative forcing. Therefore, additional ozone produced downwind of thunderstorm events is particularly effective in climate forcing (Bucsela et al., 2019).

The average value of LNO_x produced was estimated to be 180 ± 100 mol per lightning flash (Bucsela et al., 2019). LNO_x is thought to be the most important natural ozone precursor in the tropics because it is released in the middle to upper troposphere where the lifetime of ozone is long and its production efficiency per unit NO_x is high. In the tropics contribution from biomass burning source of NO_x (0.51 Tg N/yr) is greater than that from lightning 0.37 Tg N/yr), but lightning exerts a more significant role in interannual variability for tropospheric columns of ozone in the tropics as the ozone production efficiency is higher per unit NO_x from lightning (Murray et al., 2013). Finney et al. reported the ozone production efficiency from LNO_x to be 6.5 ± 4.7 times in comparison to surface NO_x sources (Finney et al., 2016a).

Studies suggest that simulated responses of LNO_x with temperature highly depend on lightning parameterization. Lightning flash densities can be simulated based on several parameterizations like the cloud top height approach, upward mass flux, and cloud ice flux. A 5–16% increase in lightning flashes per degree increase in global mean surface temperature

FIGURE 2
Shows BVOC emissions are sensitive to climate change, atmospheric CO_2 concentrations, and land cover changes. Isoprene (the most widely emitted BVOC) responds negatively to elevated O_3, drought, elevated CO_2, and land cover changes and positively to global warming. The red + sign/line indicates an increase and the blue−sign/line indicates a decrease. An increase in isoprene emissions would enhance surface ozone levels. Elevated ozone decreases net photosynthesis in plants and therefore decreases isoprene However, the net response of BVOC emissions in the future is uncertain due to the complexity of the physical and chemical processes involved and their incorporation into models

has been estimated in recent studies (Finney et al., 2018) and LNO_x emission climate response of +0.44 Tg N/K has been found using the cloud top height approach by Finney et al. (Finney et al., 2016a). Other models put estimates at +0.14 (using updraft mass flux within the cloud and cloud depth) and −0.55 Tg N/K (using updraft mass flux at 440 hPa; 49). The ice flux approach improves the simulation of lightning and the temporal correlations with ozone sonde measurements in the middle and upper troposphere. Studies using parameterizations based on the ice flux approach show a reduced amount of sensitivity or negative response to NO_x emissions. In the tropical upper troposphere, a reduction in ozone concentration of around 5–10% has been estimated (Finney et al., 2016b; Verma et al., 2021).

LNO_x plays a significant role in ozone chemistry and there is high confidence that LNO_x is disturbed by climate change still there is little confidence in the sign of the change due to basic uncertainties in the parameterization of lightning in the models (Fu and Tian, 2019; Verma et al., 2021).

Emission of biogenic volatile organic compounds

VOCs are important ozone precursors. Biogenic volatile organic compounds are released by vegetation with the main compounds being isoprene (2-methyl-1,3-butadiene) and monoterpenes but sesquiterpenes, alkenes, alcohols, aldehydes, and ketones are also present (Figure 2). These BVOCs undergo photooxidation and play a key role in atmospheric composition (Yáñez-Serrano et al., 2020; Weber et al., 2022). They influence the radiative forcing by controlling tropospheric ozone formation or by increasing the lifetime of methane by

suppressing its major sink, the OH radical (Thornhill et al., 2021). Racherla et al. showed that higher isoprene levels account for 50–60% of the increased summertime ozone episodes in the future climate simulation in the eastern United States (Racherla and Adams, 2008). The production of secondary aerosols like sulphate and nitrate particles whose formation rates depend upon the availability of oxidants is influenced by BVOCs (Unger, 2014).

The emission of BVOCs from plants is species-specific and non-linearly dependent on temperature, sunlight, soil moisture, leaf physiology, atmospheric CO_2 mixing ratio, and additional environmental variables (Ainsworth et al., 2012; Calfapietra et al., 2013; Trowbridge et al., 2020). Several species of trees in urban environments like pine, conifer, oak, eucalyptus, poplar, acacia and others emit a range of BVOCs. Aydin et al. reported the normalized total BVOC emission rate for oriental plane and sweet chestnut to be 27.1 μg/g h and 14.9 μg/g h respectively (Aydin et al., 2014).

Temperature is an important factor controlling BVOC emissions (Fitzky et al., 2019; Yáñez-Serrano et al., 2020). Temperature changes directly affect the biochemical reactions in the metabolic pathways which produce BVOCs or have indirect effects like lengthening of the growing season (Peñuelas and Staudt, 2010; Yu et al., 2021). As temperatures increase, the enzymatic activities of synthesis are enhanced, BVOCs vapor pressure is increased and the resistance of the diffusion pathway is lowered. Exponential enhancements of biogenic isoprene and monoterpene emissions with increasing temperature have been reported in several studies (Peñuelas and Staudt, 2010). Zanis et al. observed ozone enhancement over regions close to anthropogenic pollution sources or close to enhanced natural biogenic volatile organic compounds emission sources with a rate ranging regionally from 0.2 to 2 ppbv/°C due to global warming (Zanis et al., 2022). At higher temperatures the rate of emissions of significant ozone precursors produced from vegetation, like isoprene, increases significantly, doubling from 20°C to 30°C for isoprene (Archibald et al., 2020b). In the northern mid-latitudes, a modeling study reported that a 3 K temperature rise in BVOC emissions would enhance biogenic isoprene emissions by 6–31% and surface ozone by > 2 ppbv (Doherty et al., 2013). Liu et al. reported that since 2013 an increase in temperature and biogenic emissions led to a rise in the Maximum Daily Average 8-hourly (MDA8) O_3 mixing ratio by 1–2 ppbv (Liu and Wang, 2020). However, extreme temperatures and severe heat stress can cause cellular lesions and a drop in photosynthesis that may not improve upon return to cooler temperatures and reduce BVOCs emission (Yu et al., 2021).

As concentrations of CO_2 and other air pollutants are expected to rise in the future studies have estimated their impact on BVOC emissions (Sporre et al., 2019; Thornhill et al., 2021). The factors affecting these emissions are complex and interrelated like the chemical composition of the compound(s), the exposure, the concentration of pollutants, and the plant species (Yáñez-Serrano et al., 2020). The earlier studies mostly incorporated leaf-level temperature and solar radiation sensitivities and predicted a 2–3°C rise in the mean global temperature leading to an increase in global BVOC emissions by an additional 30–45% increase as a result of climate warming (Peñuelas and Staudt, 2010; Jiang et al., 2018). New studies taking into account the complex interactions between climate and ozone concentrations are challenging this pattern of much higher BVOC emissions (Unger, 2014; Harper and Unger, 2018). There is a consensus that an increase in CO_2 concentration will induce a reduction of isoprenoid emissions at least at the leaf level. On the contrary, a rise in O_3 concentration can have opposite effects depending on the extent and level of exposure—with a general stimulation in the cases of acute exposures and a general inhibition under chronic exposures (Fitzky et al., 2019).

Feng et al. examined the effect of warming, drought, elevated CO_2, and O_3 on isoprene and monoterpenes emissions. Isoprene responded negatively to elevated O_3 (−8%), drought (−15%) elevated CO_2 (−23%) but positively to warming (+53%). Effects on monoterpene emissions were lesser in comparison, significantly caused by warming (+39%; Feng et al., 2019). Faubert et al. have reported doubled volatile organic compound emissions from subarctic tundra under simulated climate warming. The relatively low emissions of monoterpenes and sesquiterpenes were doubled in response to an air temperature increment of only 1.9–2.5°C (Faubert et al., 2010).

Drought situations also hinder isoprene emissions as declining water content reduces the photosynthetic rate and stomatal conductance. Jiang et al. in a study assessing the effect of drought stress on isoprene emissions estimated the reduction of biogenic isoprene emissions by 17% globally in comparison to the non-drought simulations (Jiang et al., 2018).

Land use and land-cover changes worldwide perturb the Earth's system in numerous ways and affect BVOC emissions (Unger, 2014; Harper and Unger, 2018; Wang et al., 2021a). Squire et al. in a simulation study for the year 2095 projected a total 55% reduction in isoprene emissions in contrast with the year 2000 (Squire et al., 2014). The individual contributions from climate change, land use change (mainly cropland expansion), and CO_2 inhibition were estimated to be +30%, −73%, and − 57% respectively.

Some recent modeling studies agree that global isoprene emissions have declined since the preindustrial times and present-day varying from 10–25% driven mostly by anthropogenic land use and land cover changes (Weber et al., 2021; IPCC Pörtner et al., 2022). Climate change with warming and resultant changes in precipitation may cause a shift of vegetational zones to the north. Consequently, the shifting of woody shrubs may occur in tundra

ecosystems. Invasive insect herbivores may attack forests at higher rates increasing the local BVOC emissions (Yu et al., 2021).

The uncertainties associated with BVOC emissions are due to the complexity of physical and chemical processes mainly atmospheric CO_2, foliage composition, cover changes in natural and managed land, and their inclusion in different models (Wang et al., 2021a; Weber et al., 2022). In summary BVOC emissions will be sensitive to future climate change, land use patterns, and other environmental stressors for vegetation.

To improve understanding of BVOC emissions observational approaches have been established including indirect methods such as the application of the variability in satellite formaldehyde (HCHO) concentrations. HCHO is strongly influenced by the photochemical oxidation of VOCs and is closely linked to ozone formation. Good consistencies between satellite-based observations and the calculated photochemical property parameters like ozone forming potential of VOCs suggest that satellite-based HCHO columns can infer the VOCs emissions over large spatial areas in the future (Hong et al., 2021). Therefore, HCHO has the feasibility of being a proxy for VOCs. Simulations of BVOC emissions can be conducted using inversion studies based on formaldehyde columns observed from space using an ozone monitoring instrument. HCHO observations offer useful insights into reactive hydrocarbon emissions and the chemistry and transport model can be used to evaluate the time-dependent HCHO yield from BVOCs such as isoprene, 2-methyl-1-butanol, and α- and β-pinenes (Cai et al., 2021).

Wetland methane emissions

CH_4 besides being an important precursor of ozone in remote areas is a potent greenhouse gas. Global levels of methane have risen to 1866 ppb in 2019 from 1803 ppb in 2011, largely due to anthropogenic influence. CH_4 is the second largest contributor to climate warming and has resulted in about 21% of the increased radiative forcing since pre-industrial times. Wetlands are the biggest natural source of CH_4 which contribute between 30 and 40% to the total CH_4 emissions and are a major area of concern concerning climate change (Gedney et al., 2019; Koffi et al., 2020; IPCC Pörtner et al., 2022).

Microbial methanogenesis is a process of anaerobic respiration that occurs in water-saturated soils with limited oxygen availability and produces methane. Under this environmental condition, methane production depends on temperature, water-table depth, and both the quality and quantity of organic matter. As temperatures increase worldwide due to global warming, they may reach the optimum range for methanogenesis in more regions of the world. A rise in precipitation is also expected to be about 7%

per degree of warming (Trenberth et al., 2013). With the thawing of permafrost and local variations in the soil water content due to increased precipitation, an increase in wetland areas is expected. Annual global CH_4 emissions, in the models, differ by ±40% of the all-model mean [190 Tg CH_4/yr; 82)]. Koffi et al. have reported a rise in wetland CH_4 emissions by 50–150% during the 21st century (Koffi et al., 2020).

Elevated CO_2 will increase global, tropical, and extratropical CH_4 emissions. Higher CO_2 permits plants to increase stomatal closure which lessens water loss by the plant. This reduced water loss diminishes water demand from the soil, increasing soil moisture and thus growing wetland extent (Melton et al., 2013). Increased CO_2 in the atmosphere leads to the CO_2 fertilisation effect which finally results in enlarged methane emissions. Observations have shown more plant photosynthates are available to soil microbial communities which reduce carbon to CH_4. For doubled CO_2 an annual rise of 78% from 156 to 277 Tg/yr, in wetland methane emissions was estimated by Shindell et al. (Shindell et al., 2004).

Not only are wetland CH_4 emissions climate-sensitive but they also may provide large feedback to climate change by radiative forcing. Zhang et al. reported in comparison to anthropogenic CH_4 emissions, wetland emissions account for 14.6–25.1% of the total projected radiative forcing change which will result in added global mean radiative forcing of 0.04 W/m² to 0.19 W/m² by the end of the 21st century (Zhang et al., 2017). Gedney et al. estimated that over the 21st century wetland feedback resulted in an added temperature increase between 0.6% and 5.5% feedback on climate ranging from 0.01 to 0.11 W/m²/K (Gedney et al., 2019).

However large uncertainties are associated with wetland response to projected climate change due to limited knowledge and representation of wetland geochemistry, hydrology, and permafrost dynamics in different models. Considerable uncertainties are also associated with the assessment of natural sources and the ensuing climate feedback. Further work is required to better parameterize and evaluate the models.

Open biomass burning emissions wildfires

Wildfires emit large quantities of ozone precursors CO, NO_x, and numerous VOCs in the lower atmosphere. Jaffe et al. estimated global wildfire production of ozone to be around 170 Tg/yr, which is 3.5% of all global tropospheric O_3 production (Jaffe and Wigder, 2012). Wildfires as well play an important role in numerous atmospheric chemistry-climate feedback mechanisms.

Climate change is a significant factor in explaining variations in fire incidence and intensity in recent decades and is likely to increase the global risk of wildfires in the future (Jaffe and Wigder, 2012; Mcclure and Jaffe, 2018). Wildfire occurrence

depends on several factors like temperature, soil moisture, and the presence of trees, shrubs, and other possible fuels. Climate change is creating warmer, drier conditions. Increased drought and a lengthier fire season are enhancing these intensifications in wildfire risks (Lindaas et al., 2017; Dupuy et al., 2022).

Under the scenarios with the highest greenhouse gas emissions, Dupuy et al. in southern Europe estimated an increase in future fire, on average, from 2 to 4% (Dupuy et al., 2022). Hurteau et al. taking into consideration vegetation fire feedback and interactions of preceding fires and climate on fuel availability and flammability reported a projected increase in area burned by 14.3% (Hurteau et al., 2019).

Although O_3 production requires both NO_x and NMVOCs, wildfires tend to be NO_x-limited. McClure et al. investigated the role of wildfire emissions on O_3 production and reported MDA8 O_3 mixing ratios to be 32% higher than on non-smoky days (Mcclure and Jaffe, 2018). Fire emissions of ozone precursors vary considerably among various types of ecosystems, biomass nitrogen loads, and combustion efficiency (Liu et al., 2022). Lu et al. reported in their study that around one-third of the summer days (1989–2010) with MDA8 ozone exceeding 70 ppbv would not arise in the absence of wildfires (Lu et al., 2016). It was reported by Lin et al. that monthly mean O_3 levels could enhance by 2–8 ppb in summers due to wildfires (Lin et al., 2016). On the contrary Bourgeois et al. suggested stable biomass burning emissions since pre-industrial times (Bourgeois et al., 2021). Arora et al. showed a reduction in wildfire emissions and global areas burned due to deforestation and changes in land use (Arora and Melton, 2018).

Nevertheless, there are rising indications that wildfire emissions will play a significant role in the future composition of the atmosphere due to extended fire seasons, more intense fire activity, and a reduction of emissions from fossil fuel combustion (Burke et al., 2021). At present quantification of ozone production in wildfire plumes are difficult due to various uncertainties in measuring wildfire emissions, chemical reactions, fire vegetation feedback, and human-related factors (Dupuy et al., 2022). Accurate representation of emissions and related atmospheric chemistry in global models will be crucial to predicting future levels of surface O_3 and its role in the radiative forcing of climate and air quality.

Effect of climate change on transport patterns of ozone and its precursors

Ozone and its precursors are transported from their emission site to other downwind regions on regional, intercontinental and hemispheric scales. The pollution plumes (anthropogenic and biomass burning) can circle the globe and travel from East Asia to North America (Monks et al., 2015). Future climate change may

influence the role of long-range transport downwind effects. Several of the crucial impacts of climate change include dynamic mechanisms of the climate system and their response to anthropogenic climate forcing.

Stratosphere-troposphere exchange (STE)

The net stratospheric influx of ozone results from the stratospheric-tropospheric exchange (STE) and is comprised of troposphere-to-stratosphere transport (TST) and stratosphere-to-troposphere transport (STT). STE is impelled by the large-scale stratospheric meridional circulation known as Brewer-Dobson circulation (BDC; Archibald et al., 2020a). In the tropics air rises from the troposphere to the stratosphere, is transported to the extratropical stratosphere, and then descends from the stratosphere to the troposphere at high and middle latitudes. The role of STE in modulating tropospheric ozone is about 10% of the annual global tropospheric ozone chemical production. BDC is expected to intensify in a warming future and is estimated to lead to a 3% increase in surface ozone and a 6% increase in mid-tropospheric ozone (Hess et al., 2015). Muel et al. have projected a 53% increase in the global mean annual influx of stratospheric ozone into the troposphere between the years 2000 and 2,100 (Meul et al., 2018). Abalos et al. have reported a 10–16% increase in stratospheric ozone in the troposphere by the end of the 21st century due to the strengthening of the BDC (Abalos et al., 2020).

Tropopause folds are a key mechanism for stratospheric intrusions (SIs) in STT events. The air from SIs has high O_3, low CO, and low water vapour, in comparison to their concentrations in the troposphere. This downward transport leads to changes in tropospheric ozone levels (Xiong et al., 2022). An increase in tropospheric O_3 level is observed during the folding events, which have a stratospheric origin (Akritidis et al., 2022). Akriditis et al. identified changes in tropopause fold frequencies from 1970–1999 to 2070–2099 in both hemispheres, regionally exceeding 3%, and found links with the projected changes in the position and intensity of the subtropical jet streams (Akritidis et al., 2019). The strengthening of ozone STT in the future in both hemispheres has also been projected, with an induced surge in transported stratospheric ozone tracer through the complete troposphere, reaching up to 3 nmol/mol near the surface to 10 nmol/mol in the upper troposphere (Akritidis et al., 2019). Olsen et al. observed that ENSO-related STE related to tropopause folding in the subtropical westerly jet due to ENSO-driven STE leads to 15% of the ozone change in the NH middle troposphere near the jet in the Pacific. The variance described was more under the SH Pacific subtropical jet, with over 35% of the variance

explained by ENSO in the middle to upper troposphere (Olsen et al., 2019).

Kumar et al. observed a two-time enhancement in the surface ozone levels due to tropospheric folding facilitating STE processes over the Himalayan region. The increase in surface ozone was linked with the breaking Rossby waves in the upper troposphere (Kumar et al., 2020). Kuchar et al. associated strong orographic gravity wave drag events with increased upward Rossby wave propagation in the stratosphere. They illustrated that the drag events had the potential to alter ozone variability through changes of mixing in the surf zone and advection from lower latitudes (Kuchar et al., 2022).

Studies also indicate tropical cyclones lead to intrusion of stratospheric air into the troposphere. The tropospheric stability is weakened due to overshooting convection associated with tropical cyclones and plays a key role in STE (Shen et al., 2015; Ratnam et al., 2016). Das et al. observed an increase in surface ozone levels by ~ 10 ppbv in the daytime and 10–15 ppbv in the night-time during a cyclone. They also report the descent rate of the enhanced ozone layer during the passage of the tropical cyclone to be $0.8–1 \, km \, day^{-1}$, which is three times that of a clear-sky day. The increase in the upper-tropospheric (10–16 km) ozone was 20–50 ppbv, which stretched down to the middle (6–10 km) and lower troposphere (<6 km; (Shankar Das et al., 2016)).

Knowland et al. examined the influence of mid-latitude cyclones on European background surface ozone influence. They identified the importance of the passage of a cyclone's cold front, the ability of cyclones to bring down high levels of O_3 from the stratosphere, and associated surface high-pressure systems as the main drivers associated with cyclones that impact surface ozone (Emma Knowland et al., 2017).

Long-range transport of ozone

Long-range transport of ozone is affected by ENSO, the Arctic Oscillation (AO), and the closely related North Atlantic Oscillation (NAO) which impact weather patterns in North America and Eurasia. It also includes the Asian summer monsoon upon which more than a billion people depend for their freshwater supply.

The El Niño–Southern Oscillation (ENSO)

ENSO refers to the interaction between the atmosphere and ocean in the tropical Pacific and is a cause of large-scale climate variability in the coupled atmosphere-ocean system (Cagnazzo et al., 2009). It affects severe weather (impacting western United States drought and Atlantic hurricane activity), rainfall, river flow, and agricultural production over different parts of the world (Lin and Qian, 2019). It is a critical climate occurrence that can change atmospheric circulation across the globe and changes tropospheric ozone on an interannual timescale (Oman et al., 2013; Nowack et al., 2017). ENSO-related changes in circulation, thermal patterns, and composition drive tropical ozone variability (Oman et al., 2011). Oman et al. have reported the correlation of the measurement-derived Ozone ENSO Index with ENSO to be 0.84. Yang et al. reported a positive correlation with the ENSO index of summertime ozone concentrations in China in the years 1990–2019, with increases by 20% over southern China in El Niño relative to La Niña years (Yang et al., 2022).

Rowlinson et al. observed ENSO influence on fire incidence, wetland emission, and atmospheric circulation-affected sources and sinks of tropospheric O_3. They reported greater fire emission of precursors like NO_x and CO-enhanced O_3 which led to an O_3 radiative effect of 0.03 W/m^2 (Rowlinson et al., 2019). Xue et al. observed that ENSO-modulated fires in Southeast Asia, dominate the interannual variability of springtime trans-Pacific transport of ozone across the entire North Pacific Ocean. During El Nino springs, the intensified fires from Indochinese Peninsula and Indonesia, together with large-scale circulation anomalies, resulted in enhanced ozone plumes stretching over 15000 km in both the lower-middle and upper troposphere (Xue et al., 2021). This enhancement is also observed in the *in situ* measurements of ozone concentration, with an almost 10% increase at Mauna Loa Observatory, Hawaii, the long-distance transport over the North Pacific (Xue et al., 2021). Peiro et al. described the significant impact of the El Niño event on the tropospheric column ozone, with 20% higher values in Indonesia with the Indian Ocean region and 10% lower in the Pacific Ocean center region (Peiro et al., 2018).

Warmer and drier weather conditions during El Niño also promote biomass burning there and may increase lightning activity both contributing to higher ozone (Zhang et al., 2011). It has been reported that both strong La Niña and strong El Niño events lead to the greater entry of water vapor (0.5 ppmv increase, about 70% of observed increase) and stratospheric moistening in the tropics (Garfinkel et al., 2018). This may lead to greater ozone loss through HO_x chemistry. Shen et al. studied the effect of El Niño on MDA8 ozone concentrations in summer over the eastern United States from 1980 to 2016 and reported each standard deviation rise in the Niño 1 + 2 index was connected to an increase of 1-2 ppbv ozone in the Atlantic states and a decrease of 0.5-2 ppbv ozone in the south-central states (Shen and Mickley, 2017). Rowlinson et al. estimated El Niño-related variations in atmospheric transport and humidity diminished global tropospheric O_3 concentrations which resulted in a −0.03 W/m^2 change in the O_3 radiative effect (Rowlinson et al., 2019).

Nowack et al. have highlighted the role of ozone feedback in the ENSO-ozone response under the scenario of global warming. Lapse rate changes due to ozone feedback may modify the

Walker circulation and accordingly, tropical Pacific surface temperature gradients. Ozone feedback diminishes the slowing of the Walker circulation under CO_2 forcing (Nowack et al., 2017). ENSO is also cognized to control the strength and frequency of STT of ozone over the Pacific-North American region (Albers et al., 2022). It has been reported that interannual variations in the strength of the ENSO of around 40% affect the STT flux causing variations in tropospheric ozone at northern midlatitudes of around 2% (Archibald et al., 2020a).

Yang et al., and Xue et al. found a positive correlation between ozone concentrations and the ENSO index. Shen et al., Oman et al. and Peiro et al. reported that ozone levels varied according to the phase of ENSO and had different effects in different regions. Hope et al. suggest that external forcing plays a role in the ENSO variance changes (Hope et al., 2017) while Maher et al. do not find external forcings to modulate ENSO over historical periods (Maher et al., 2018). Although there is high confidence that ozone is sensitive to the strength of ENSO forcings in the troposphere, there is low confidence that climate change influences the observed changes in ENSO.

Arctic Oscillation and North Atlantic Oscillation

AO and NAO are leading modes of climate variability in the northern hemisphere and influence weather patterns in North America and Eurasia (Mann, 2021). In the present climate, indices of the AO and NAO are highly correlated in time, due to a constant mechanism influencing both-the downward propagation of stratospheric anomalies into the troposphere.

Pollution transport pathways lead to the accumulation of ozone precursors in the northern hemisphere troposphere in winter and their release in lower latitudes in spring (Eckhardt et al., 2003). AO and NAO influence tropospheric ozone pollution at mid-latitudes. NAO is reported to cause prominent surface ozone anomalies in the lower troposphere in Northern Europe due to the increased flow of westerlies from the United States to Europe in the positive NAO phase (Russo et al., 2022). NAO affects surface ozone concentrations on a monthly timescale, over 10 ppbv in southwestern, central, and northern Europe during all seasons except fall (Pausata et al., 2012). It has been suggested that the vertical distribution of ozone in the stratosphere in the northern hemisphere can be affected by AO (Liu and Hu, 2021). A 72% of the variance in the total ozone column in the high northern latitudes was attributed to AO (Sekiya and Sudo, 2012). In North Atlantic, the variability of upper tropospheric ozone is very strongly influenced by the STE and tropical lightning NO_x emissions which are in turn influenced by AO/NAO (Russo et al., 2022).

Studies state that tropospheric climate changes impact both the AO and NAO and the effects are spread throughout the troposphere. The results displayed by the experiments with

tropospheric warming or stratospheric cooling produce give positive AO/NAO indices (Liu and Hu, 2021).

During high NAO phases, increased transport of pollutants to the Arctic was observed from NH, especially in winter and spring. A positive trend in NAO and enhanced transport of pollutants including ozone into the Arctic may be partly responsible for rapid climate change (Eckhardt et al., 2003).

The longitudinal response of ozone to the AO in the upper troposphere is controlled by the variations in tropopause height connected with the AO, such that lower (higher) heights imply increased (decreased) ozone. AO can modulate ozone by up to 5 ppbv (Hess and Lamarque, 2007). In Northern Canada and the Arctic, AO is correlated with ozone decreases as variations in stratospheric ozone dominates the ozone signal throughout the depth of the troposphere. In other locations, the local response to the AO is governed by changes in the transport of ozone produced in the troposphere. The local response to the AO is also dictated by fluctuations in the transport of ozone produced in the troposphere. This occurs at locations over the lower troposphere over the Atlantic basin where ozone is negatively correlated with the AO, and at locations over the lower troposphere over Europe and Eastern Siberia where ozone is positively correlated with the AO.

Most climate models project an increase in the winter NAO index most likely in response to climate change (Gillett et al., 2003; Rind et al., 2005). At the beginning and end of the 20th century, an increase in ozone over Greenland and/or its depletion over the Azores was accompanied by a positive NAO index. In contrast, the negative NAO phase, which was observed in the middle of the 20th century, was attributed to the O_3 reduction at polar latitudes (Kilifarska et al., 2020).

Hamounda et al. reported a breakdown of the coupling between the AO and the NAO in the warm climate projections and projected changes in mean atmospheric circulation (Hamouda et al., 2021). It has also been reported that the inclusion of ozone photochemistry is necessary to capture key regional climate responses in models simulating AO/NAO patterns highlighting ozone-AO connections (Mann, 2021).

Synoptic scale meteorology

In polluted areas, high concentrations of tropospheric ozone have often been correlated with meteorological variables, like high temperature, strong solar radiation, low relative humidity, low precipitation, and poor meteorological transport and mixing on synoptic and seasonal timescales. Meteorological conditions play a vital role in controlling ozone concentrations, not only through transport but also by affecting natural emissions and chemical rates (Li et al., 2020). Yang et al. have reported that variations in meteorological conditions caused up to 15% variation in annual ozone concentrations in some regions in

China (Yang et al., 2019). Cheng et al. suggest that the relative contribution of meteorological conditions to long-term variation in ozone in Beijing is only 2%–3%, but short-term ozone concentrations were affected significantly by variations in meteorological conditions (Cheng et al., 2019). The appropriate NO_x/VOC reduction ratio may also differ significantly under variable meteorological conditions (Yang et al., 2019).

These associations suggest that changes in regional meteorology due to climate change will possibly incur changes in future tropospheric ozone. Several such synoptic and seasonal-scale weather patterns have been recognized as partial drivers for high tropospheric ozone, and the likely changes in some of those weather patterns in future climates have been explored.

O_3 is strongly correlated with temperature due to associations of increased temperature with enhanced O_3 photochemical reaction rates mostly driven by an increase in the rate coefficients for chemical processes, which results in the presence of positive activation energy barriers for chemical reactions (Jacob and Winner, 2009). Temperature also affects PAN decomposition, SNO_x emissions, and BVOC emissions as discussed earlier. O_3-temperature correlation also depends on its variation with other meteorological phenomena like stagnancy and humidity (Porter and Heald, 2019). An increase in temperature may also enable different pathways which may result in a greater tendency for producing ozone. For example, the rate constant for the reaction between OH and methane, is strongly temperature dependent and increases by a factor of 16 when the temperature rises from 200 to 300 K (Archibald et al., 2020b). This offers a globally important source of peroxy radicals (RO_2) which mediate the formation of tropospheric ozone. Gu et al. observed O_3 concentrations to increase faster with rising temperatures at mean rates of 6.65 and 13.68 ppb/°C at an urban and remote site in China respectively (Gu et al., 2020).

With the ongoing rise in greenhouse gases and corresponding increases in average temperature, heat waves are expected to become more intense over global land areas. High temperatures are frequently associated with higher surface ozone and its related negative impacts (Zhao et al., 2018; Li et al., 2020; Zhan et al., 2021). Meehl et al. reported that if ozone precursors emissions remain the same then, surface ozone concentrations increase on future heat wave days compared to non-heat wave days in most regions of north-eastern North America, western Europe, central and northern Asia but decreases in some regions like coastal and south-eastern North America, Eastern Europe, north-eastern Asia where there is ozone suppression likely due to isoprene emissions (Meehl et al., 2018). But emissions of ozone precursors will change in different emission scenarios and an increase in temperature will affect VOC emission and thus ozone concentrations (Orru et al., 2019). Therefore, temperature is not a key control of modeled ozone concentrations in future heat waves (Archibald et al., 2020b).

According to the sixth assessment report of the Intergovernmental Panel for Climate Change (IPCC), it is likely that human influence has contributed to moisture in the upper troposphere since 1979. Also, there is medium confidence that human influence contributed to a global increase in annual surface-specific humidity, and medium confidence that it contributed to a decrease in surface relative humidity over midlatitude NH continents during summertime. Regional O_3 pollution episodes occurred frequently when humidity is low (Zhan and Xie, 2022) and an increase in water vapor can lead to a decrease in ozone concentrations in the lower troposphere (Liu and Wang, 2020). Zanis et al. report climate change benefits on surface ozone of −0.96 ± 0.07 ppbv/°C which is mainly linked to the dominating role of enhanced ozone destruction with higher water vapour abundances under a warmer climate (Zanis et al., 2022). In a study by Qu et al., it has been stated that reduced water vapour caused fewer clouds formation resulting in more sunlight reaching the surface, so the photolytic production of ozone increased. Due to the collective effect of the two meteorology feedback systems, the annual average ozone concentration in China declined by 2.01 ppb (6.2%; (Qu et al., 2021)).

Mid to high-latitude land precipitation has increased over the northern hemisphere, and the zonal mean wet tropics dry subtropics contrast has been strengthened due to human influence (IPCC Pörtner et al., 2022). Precipitation decreases the ozone concentration via the wet removal of ozone precursors (Liu and Wang, 2020). Wind fields can significantly affect ozone by transporting ozone and ozone precursors in and out of different regions (Liu and Wang, 2020). Distinct seasonal transitions in prevailing wind and rainfall associated with the monsoon circulation result in unique ozone variations in many regions of the world (Lu et al., 2018). Studies report that the onset of the summer monsoon caused ozone-unfavourable meteorological conditions and strong upward transport, which all led to large decreases in the lower tropospheric ozone burden in India. Rainfalls efficiently removed ozone, and weak solar radiation combined with low temperatures resulted in low ozone photochemical formation. In addition, the presence of water vapor from marine air resulted in the chemical loss of ozone at low NO_x conditions along with negligible biomass burning and low anthropogenic emissions led to a reduction in monthly ozone chemical production by 4.2 Tg (from 11.8 Tg in May to 7.6 Tg in August) as integrated over the Indian terrestrial land (Lu et al., 2018).

Similar ozone-monsoon responses but with different seasonal variations were also found for near-surface ozone in China (Lu et al., 2019). Interannual ozone variability in monsoon regions shows strong correlations with monsoon strength. Sometimes due to varied reasons, a negative correlation between ozone levels and monsoon strength has been reported in different sites in the Asian continent due to

stronger net chemical production (Wang et al., 2021b). Yang et al. reported changes in the East Asian summer monsoon strength caused variations in O_3 concentrations over central-eastern China by 2%–5%, ascribed to the changing transboundary transport of O_3, with high O_3 levels appearing in strong monsoon years (Yang et al., 2014).

Climate variabilities of ENSO also affect Asian summer monsoon (ASM) anticyclone circulations and may influence tropospheric ozone through changes in monsoon on a longer timescale. Kunze et al. observed a weakening of the ASM during ENSO warm events (Kunze et al., 2016). The Asian summer monsoon (ASM) anticyclone plays a vital role in the efficient transport of Asian tropospheric air masses to the extratropical upper troposphere and lower stratosphere (UTLS). Park et al. observed minima in ozone within the anticyclone in the UTLS (Park et al., 2007). Randel et al. observed that ASM anticyclone linked to transient convective events was associated with the vertical transport of low ozone and high-water vapor into the UTLS region (Randel and Park, 2006). Fadnavis et al. assessed that eddy detached from the anticyclone resulted in the transport of Asian trace gases away from the Asian region to the western Pacific and western Africa and a 10% reduction of NMVOCs and NO_x Asian emissions caused a decrease in peroxyacetyl nitrate (2%–10% near 200–80 hPa), ozone (1%–4.5% near ~ 150 hPa) and ozone heating rates (0.001–0.004 K/day near 300–150 hPa) in the upper troposphere over western Africa and the western Pacific (Fadnavis et al., 2018). Li et al. reported ozone variations in over Lhasa to be associated with a combination of processes, namely the intrusion of ozone-rich stratospheric air, the upward transport of low ozone concentrations in the marine boundary layer by tropical cyclones, and the uplift of polluted air rich in ozone in South Asia (Li et al., 2018).

Extratropical jets are wind maxima in the upper troposphere, often accompanied by storms, blocking, and weather extremes. Blocking refers to long-lived, stationary high-pressure systems that are often linked with a poleward displacement of the jet, producing cold spells in winter and heat waves in summer. Das et al. have reported an increase in ozone concentration in the upper troposphere with the strength of the tropical easterly jet, which is a consequence of ASM circulation (Das et al., 2019). IPCC's sixth assessment report states a likely increase in the total number of extratropical cyclones since the 1980s in the Northern Hemisphere (NH) but a reduction in deep cyclones in summer. The reduction in cyclone activity by about 4% per decade NH in summer is likely related to human-induced warming. Boreal winter witnessed the potential influence of the rapid Arctic warming on the observed intensification of NH storm track activity in the past few decades (IPCC Pörtner et al., 2022).

Recently some studies suggest that the oxidation of soot particles with ozone increases their ability to act as cloud condensation nuclei (CCN) and is an important pathway to be considered in future studies (Carnerero et al., 2019; Friebel and Mensah, 2019). Friebal et al. report an increase in the number of CCN due to O_3 aging of soot can lead to an increase of the CCN burden on a global scale (+12.7%) and a significant enhancement of the cloud droplet number concentration in northern midlatitudes (up to +30.3%). Reaction temperature also has a pronounced effect on CCN activation of soot particles and combined with high ozone levels may have a significant impact on new particle formation.

Plausible changes in temperature and ozone in the future

What the future holds in terms of ozone concentrations will be decided by different factors including government policies and the choices that the world makes today. The Shared Socioeconomic Pathways (SSPs) furnish a framework for combining future climate scenarios with socioeconomic development and provide elaborate information about plausible future evolutions of society and natural systems (Nazarenko et al., 2022). In all SSP, global temperatures will continue to rise for at least a few decades (Figure 3).

A warmer climate has the potential to increase surface ozone concentrations and this is referred to as the ozone climate penalty. This penalty effect will be most likely in polluted regions (Bloomer et al., 2009; Finney et al., 2016b; Nazarenko et al., 2022). Gong et al. have observed that different climate scenarios have penalty effects on summertime ozone concentrations of about 4–10 ppbv in the eastern United States, 1-5 ppbv in Southern Europe, and 3-6 ppbv in Northeastern Asia in the 2100s compared to the 2000s.

In the SSP1-2.6 scenario, surface ozone may decline in many parts of the world but in the SSP5-8.5 scenario, it is expected to significantly increase (Figures 4A–D; (Gong et al., 2022)). Gong et al. reported a reduction in O_3 exceedance levels on hot days from 3.7 days (2030) to 3.0 (2050) in SSP 1–2.6 scenario. But under the SSP 5–8.5 scenario reported co-occurrences of O_3-polluted days and hot days from 4.5 (2030) to 6.4 (2050). Shang et al. predict a significant rise in the tropospheric ozone concentrations in the middle and lower troposphere under the SSP3-7.0 and the SSP5-8.5 scenarios (Shang et al., 2021). Large regions in South and East Asia will witness enormously high levels of ozone in the future the SSP3-7.0 and the SSP5-8.5 scenarios (Zhang et al., 2022b; IPCC Pörtner et al., 2022). However, Wang et al. reported a decrease in O_3 concentrations from 2015 to 2,100 under SSP1-2.6 and SSP2-4.5 (Wang et al., 2022). Zanis et al. predicted a reduction in surface ozone under the SSP3-7.0 scenario in unpolluted regions (from −0.2 to −2 ppbv/°C) and globally (−1 ppbv/°C) as a result of accelerated ozone chemical loss due to greater water
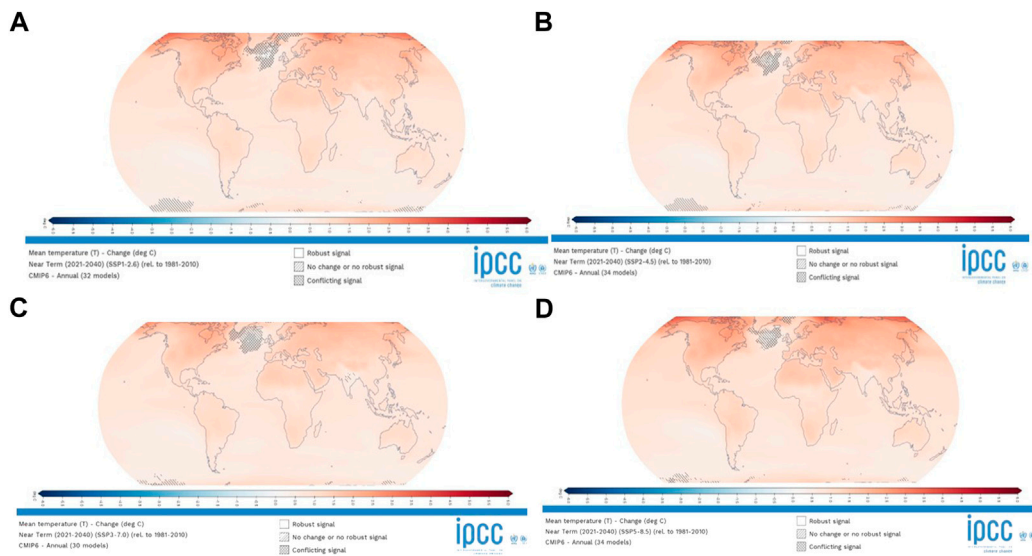
FIGURE 3
(A–D) show changes in mean temperature in different SSP scenarios in the near term (2021–2040) relative to 1981–2010 by Iturbide, M., Fernández, J., Gutiérrez, J.M., Bedia, J., Cimadevilla, E., Díez-Sierra, J., Manzanas, R., Casanueva, A., Baño-Medina, J., Milovac, J., Herrera, S., Cofiño, A.S., San Martín, D., García-Díez, M., Hauser, M., Huard, D., Yelekci, Ö. (2021) Repository supporting the implementation of FAIR principles in the IPCC-WG1 Atlas. Zenodo, DOI: 10.5281/zenodo.3691645. Available from: https://github.com/IPCC-WG1/Atlas

FIGURE 4
(A–D) show changes in ozone in different SSP scenarios in the near term (2021–2040) relative to 1981–2010 by Iturbide, M., Fernández, J., Gutiérrez, J.M., Bedia, J., Cimadevilla, E., Díez-Sierra, J., Manzanas, R., Casanueva, A., Baño-Medina, J., Milovac, J., Herrera, S., Cofiño, A.S., San Martín, D., García-Díez, M., Hauser, M., Huard, D., Yelekci, Ö. (2021) Repository supporting the implementation of FAIR principles in the IPCC-WG1 Atlas. Zenodo, DOI: 10.5281/zenodo.3691645. Available from: https://github.com/IPCC-WG1/Atlas
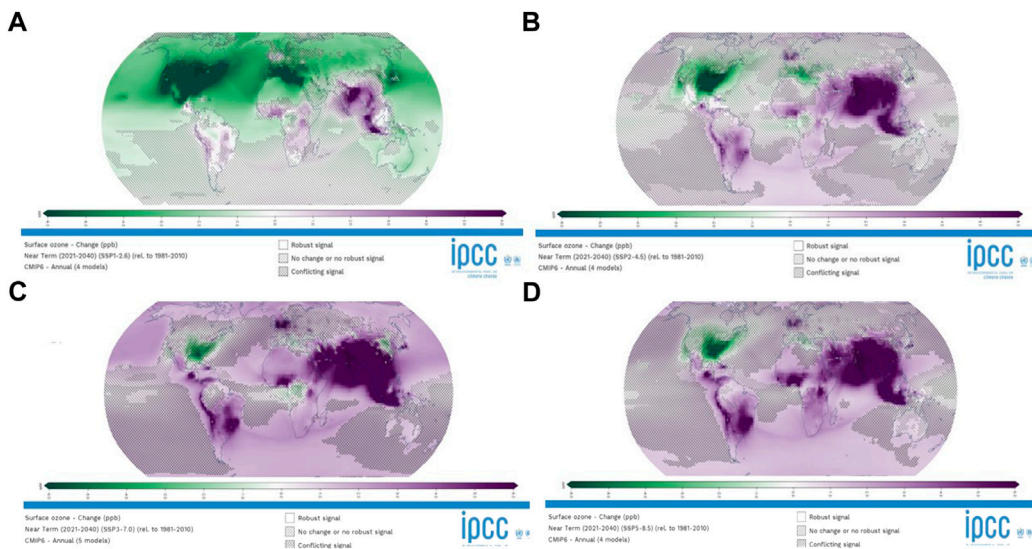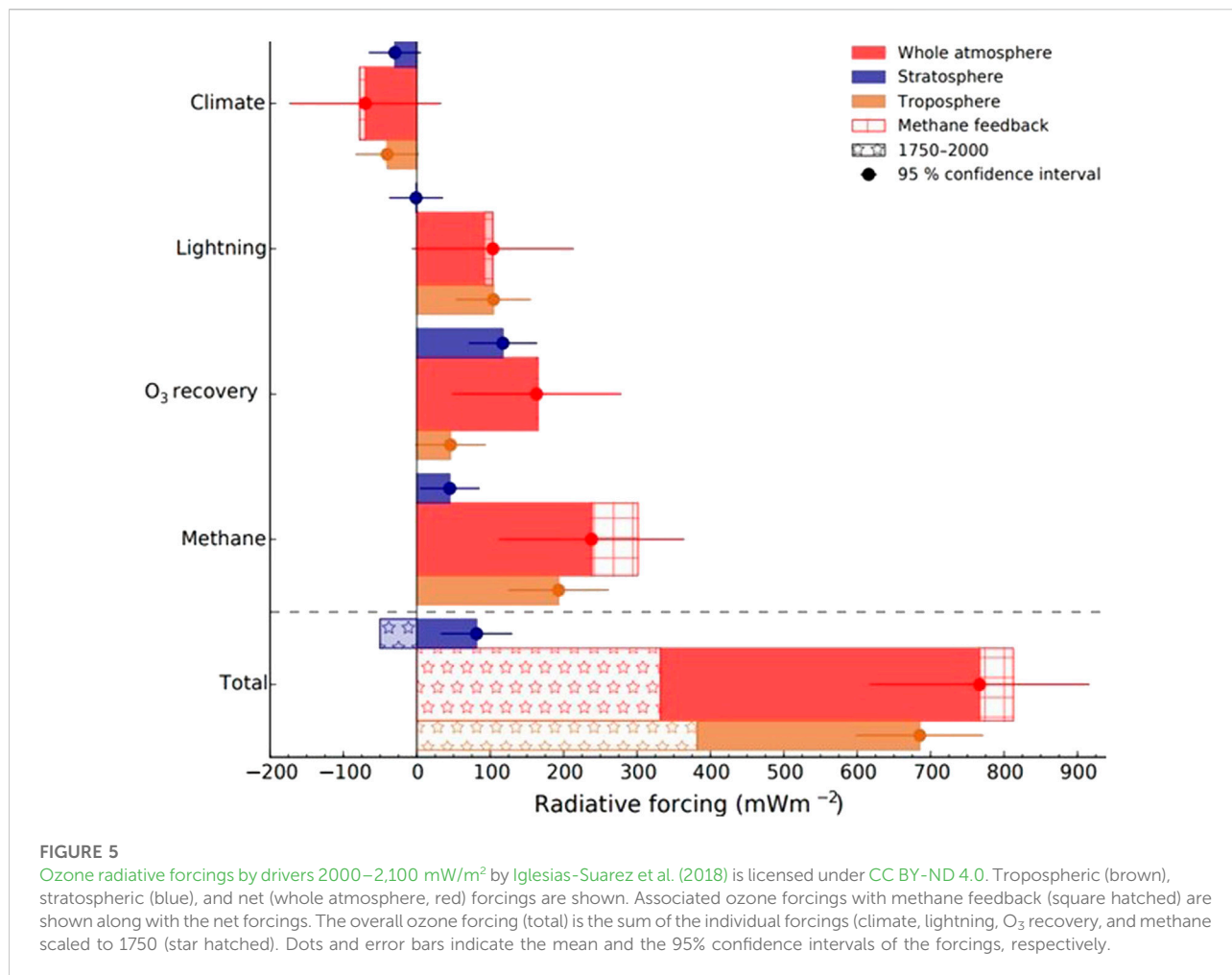
FIGURE 5
Ozone radiative forcings by drivers 2000−2,100 mW/m² by Iglesias-Suarez et al. (2018) is licensed under CC BY-ND 4.0. Tropospheric (brown), stratospheric (blue), and net (whole atmosphere, red) forcings are shown. Associated ozone forcings with methane feedback (square hatched) are shown along with the net forcings. The overall ozone forcing (total) is the sum of the individual forcings (climate, lightning, O_3 recovery, and methane scaled to 1750 (star hatched). Dots and error bars indicate the mean and the 95% confidence intervals of the forcings, respectively.

but increase in surface ozone (up to 2 ppbv/°C) over polluted regions (Zanis et al., 2022).

Radiative forcing of ozone

Ozone is the third strongest contributor to the total tropospheric radiative forcing (RF) since preindustrial times. Estimates of the pre-industrial to present-day tropospheric ozone radiative forcing are based entirely on models due to the absence of observed pre-industrial ozone measurements. Checha-Garcia et al. estimated the RF for tropospheric ozone and stratospheric ozone to be 0.33 W/m² and −0.029 W/m² respectively, giving a total ozone RF of 0.30 W/m² in 2000–2014 relative to 1850–1860 (Checa-Garcia et al., 2018). Zhao et al. have also observed that an increase in tropospheric ozone and a decrease in stratospheric ozone led to contrasting forcings. They reported the global mean 1970–2010 effective RF to be 0.24 ± 0.01 W/m² for tropospheric ozone with the largest value of 0.47 ± 0.31 W/m² over the Arctic and effective RF due to

stratospheric ozone depletion to be −0.07 ± 0.15 W/m² (Zhao et al., 2019). Skeie et al. assessed the ozone RF to be 0.35 W/m² in 2010 relative based on models with both tropospheric and stratospheric chemistry (Skeie et al., 2020).

Turnock et al. predicted a change in the global tropospheric O_3 RF of +0.07 W/m² from 2010 to 2050 based on changes in CH_4 abundance and tropospheric O_3 precursor emissions (Turnock et al., 2018). Estimates of historical and future changes in O_3 and its RF over 300 years under different SSPs have been assessed (Turnock et al., 2019). Changes in surface O_3, tropospheric O_3 burden, and O_3 radiative forcing has been predicted to be +8 ppbv, +76 Tg, and +0.3 W/m² respectively from 1750 to 2014. Turnock et al. also projected reductions in global surface O_3 concentrations of more than 3.5 ppbv and global O_3 RF of less than—0.1 W/m² in future scenarios from 2014 to 2050 in case of strong air pollution mitigation measures. An increase in O_3 across all regions and O_3 radiative forcing >+0.05 W/m² was reported with weak air pollution mitigation measures (Turnock et al., 2019). Iglesias-Suarez et al. calculated net ozone forcing in the year 2,100 to be 430 ± 130 m W/m² relative to the year 2000 and 760 ± 230 mW/m² relative to the

year 1750 (Iglesias-Suarez et al., 2018). The RFs due to climate change, ozone-depleting substances, and methane were estimated to be $33 \pm 104 \text{ mW/m}^2$, $163 \pm 109 \text{ mW/m}^2$, and $238 \pm 113 \text{ mW/m}^2$ respectively (Figure 5).

Summary and conclusion

Ozone is a radiatively active gas, which interacts with both shortwave and longwave radiation. In the troposphere, O_3 is an important secondary pollutant that regulates the oxidizing capacity of the atmosphere and is a source of OH radicals. Future changes in emissions of natural precursors of ozone will be a result of the complex interplay between its photochemistry, climate change, and transport mechanisms.

Precursors like SNO_x, LNO_x, biogenic VOCs, and wetland methane emissions are characterized by large year-to-year variations and have a strong potential of being influenced by large changes in their emissions ensuing from future climate change. SNO_x differs non-linearly with region-specific agricultural management, soil conditions, and meteorology and in drylands may predominantly be emitted as a pulsed flux in response to irrigation/precipitation-drying cycles. SNO_x is expected to increase in a warming climate but factors affecting it are not well constrained in models (Lu et al., 2021).

Lightning is a main natural source of nitrogen oxides in the troposphere with important consequences for atmospheric composition in the middle to the upper troposphere and the lower stratosphere. LNO_x emission sensitivity to climate change is in the range of 4–60%/K (Verma et al., 2021). Current studies find LNO_x emission climate sensitivity lying at the lower end of this estimate and uncertainty exists even in the sign of future projection responses due to different parameterizations (Finney et al., 2018).

BVOC emissions are affected by temperature, drought, solar radiation, humidity, availability of nutrients, CO_2, O_3, etc. Isoprene and monoterpenes are predominant VOCs released by plants. Although studies have observed a decline in isoprene emissions their impact in future years will depend on changes in land use patterns, changing climate, and atmospheric CO_2 concentrations (Cai et al., 2021).

Wetlands are the largest contributors to CH_4 emissions (Zhang et al., 2017). These emissions are highly sensitive to climate change and also provide positive feedback to climate change. However present-day estimations of their response to climate are highly uncertain (Archibald et al., 2020b).

STE is associated with synoptic-scale and mesoscale processes, such as tropopause folds, influenced by deep stratospheric intrusions, tropopause folds, gravity wave breaking, and deep convection and cyclones (Meul et al., 2018). STE is projected to increase due to climate and may lead to enhancement in surface ozone levels.

Tropospheric ozone levels are robustly correlated in a non-linear manner with temperatures, especially in highly polluted regions. It depends on temperature-dependent mechanisms related to O_3 precursor emissions, lifetimes, and reaction rates. More water vapour and higher temperatures in a warmer climate would enhance ozone chemical destruction, leading to lower baseline surface ozone levels in unpolluted regions (Li et al., 2020; Zanis et al., 2022). In polluted regions surface temperatures will trigger regional feedbacks in chemistry and local emissions that will increase surface ozone and intensity of surface O_3 peaks (Zanis et al., 2022).

Modes of climate variability like ENSO, NAO, and AO influence atmospheric circulations and weather conditions and will likely be influenced by climate change. Their impact on ozone levels differs from region to region depending on their phase of development and different indices.

Counteracting effects make the response of tropospheric ozone to climate-induced earth system changes complex (Kang et al., 2020; Zhan and Xie, 2022). Studies considering the individual effects of climate-driven changes in specific precursor emissions processes show increases in surface ozone under a warmer atmosphere for some processes. This is indeed the case for enhanced STE and SNO_x. However, quantification of emissions from natural systems is difficult to quantify in isolation due to complexities in the understanding of chemical mechanisms and processes and their incorporation into models.

Looking forward it is clear that climate and ozone levels are deeply intertwined. Climate influences ozone concentrations through several processes and is affected by some of these which provide a feedback loop and perturb the climate even more. The review examined the new studies on the interactions of climate change on the chemistry and deposition, natural precursor emissions of ozone, and long-range transport of ozone. Fluctuations in climate, land use patterns, and increasing CO_2 concentrations will influence BVOC emissions, SNO_x, LNO_x, wetland CH_4, and wildfire emissions in a complex manner but there remains considerable uncertainty in how they will impact future ozone levels. The quantification of the effects is challenging since it involves complex interactions in the atmosphere as well as the biosphere. Emission controls on ozone precursors need to be inflicted irrespective of the climate policy and this becomes imperative due to unequivocal evidence of adverse health impacts associated with these pollutants.

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AL- Conceptualization and review editing. SD- Writing original draft, visualization, reviewing, and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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