Article:
Tan, Z., Ma, X., Lu, K. et al. (2026) Governing atmospheric oxidation capacity is the key to synergistic air quality and climate gains. One Earth, 9 (1). 101569. ISSN: 2590-3330

https://doi.org/10.1016/j.oneear.2025.101569

Governing Atmospheric Oxidation Capacity as the Key to Synergistic Air Quality and Climate Gains

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SUMMARY

Effective management of air quality and climate change requires recognition of their fundamental coupling through atmospheric oxidation capacity (AOC), which governs the atmosphere's self-cleansing capacity. However, policies often overlook the nonlinear chemical feedbacks inherent to AOC, leading to fragmented strategies that risk unintended consequences. Here we demonstrate that uncoordinated strategies, such as reducing fossil fuel-related NO_X without concurrent methane controls, can suppress OH radicals, inadvertently prolonging methane's lifetime. This "chemical lockdown paradox", observed during COVID-19 lockdowns, reveals critical trade-offs, where short-term air quality gains may increase methane accumulation, offsetting the climate benefits of CO_2 abatement. Given AOC's spatial heterogeneity, such effects can extend beyond local scales. We thus propose a regulatory framework integrating AOC dynamics through coordinated multi-pollutant controls, advanced multiscale AOC monitoring, and improved Earth system models with fully-coupled chemical feedbacks. This framework paves a science-based pathway for synergistically managing air quality and climate mitigation throughout the decarbonization transition.

AOC: THE UNSEEN ATMOSPHERIC GOVERNOR

Air pollution and climate change constitute two intrinsically linked environmental crises that pose severe threats to human health and ecosystem security. This linkage stems from the fact that many anthropogenically emitted gases and particulate matter simultaneously impact both air quality and climate warming. Consequently, mitigation efforts targeting one crisis inevitably influence the other. Nevertheless, conventional policy frameworks

have predominantly emphasized targeted reductions of specific pollutants, including nitrogen oxides (NO_X), sulfur dioxide (SO_2), fine particulate matter ($PM_{2.5}$), methane (CH_4), and carbon dioxide (CO_2). However, atmospheric responses to such emission controls are often nonlinear and complex. A critical yet often overlooked factor governing these responses is the Atmospheric Oxidation Capacity (AOC), which refers to the ability of the atmosphere to cleanse itself of pollutants through chemical oxidation (Figure *1*). AOC plays a decisive role in regulating the atmospheric lifetime of potent greenhouse gases such as CH_4[1], while simultaneously determining the formation of secondary pollutants like ozone and $PM_{2.5}$[2-3], both of which are also significant climate forcers. Emission reduction strategies targeting individual pollutants without accounting for their interactive effects on AOC, risk triggering unintended atmospheric consequences that may undermine both air quality gains and climate mitigation efforts[4-5].

The COVID-19 lockdowns offered a compelling real-world illustration of this systemic risk. Rapid declines in transportation and industrial activity led to sharp reductions in NO_X emissions, which is a common target in decarbonization and clean air policies[6-7]. However, this decrease also suppressed the formation of hydroxyl radical (OH), the atmosphere's primary oxidant, leading to a decline in atmospheric cleansing capacity and consequent accumulation of CH_4[8-9]. Studies indicate that the reduction in OH levels during early 2020 contributed to a 53% increase in the methane growth rate anomaly[4], partially offsetting the climate benefits associated with reduced CO_2 emissions. This "chemical lockdown paradox" underscores a critical governance challenge: effective polices must consider not only what we remove from the atmosphere, but also how those removals affect the very processes that sustain atmospheric health. If CH_4 accumulation outpaces AOC

recovery, a destabilizing “runaway” effects could occur[10], potentially collapsing the atmospheric self-cleansing capacity.

Analogous trade-offs are evident in regional air quality management. For example, stringent NO_X reduction in the absence of concurrent controls on volatile organic compounds (VOCs) has in some urban regions inadvertently increased ozone pollution[11-12]. Likewise, efforts to reduce $PM_{2.5}$ concentrations by controlling aerosol precursors may modify photolysis rates and alter oxidative pathways, in some cases raising surface ozone levels in certain regions[13-14]. Studies suggest that a major driver of ozone increase in the North China Plain was the approximately 40% reduction in $PM_{2.5}$ between 2013-2017, which slowed the aerosol sink of hydroperoxyl radical (HO_2) and thereby accelerated ozone production[5]. These unintended consequences highlight the inherent constraints of isolated emission control strategies. To achieve synergistic improvements in both ozone and $PM_{2.5}$, policies must be designed with a holistic understanding of their combined effect on AOC.

The integration of AOC into air quality and climate governance is growing increasingly urgent amid rapidly evolving emission policies. Although global efforts to reduce fossil fuel consumption and decarbonize energy systems are essential, these initiatives may profoundly reshape the atmospheric chemical environment, notably altering the abundance of OH[22]. Effectively addressing climate change and protecting air quality therefore require a nuanced, systems-oriented approach centered on sustaining AOC. In this perspective, we systematically evaluate the key chemical drivers and regional variations of AOC, identify critical knowledge gaps in current modelling frameworks, and propose an integrated framework for monitoring and governing AOC across scales. By establishing AOC as a central scientific and policy nexus in atmospheric and climate governance, co-designed strategies can be

developed, optimized, and implemented to enhance atmospheric resilience and deliver dual environmental and climate benefits.

CURRENT UNDERSTANDING OF TROPOSPHERIC AOC

Fundamental Chemistry and Governing Mechanisms

The stability of AOC hinges on a finely tuned set of chemical reactions dominated by OH radicals (Figure *1*)[15]. These radicals are generated globally via the photolysis of ozone in the presence of water vapor. Through catalytic oxidation, OH radicals remove pollutants such as CH_4, CO, and VOCs, a process that simultaneously produces peroxy radicals (RO_2). The resulting peroxy radicals subsequently oxidize NO to NO_2, which undergoes photolysis to regenerate O_3 and recycle NO, thereby sustaining the oxidative cycle. Radical propagation terminates either through the reaction between OH and NO_2, forming nitric acid (HNO_3), or via peroxy radical recombination, which produces stable peroxides (ROOH)[16]. Notably, the very mechanisms responsible for atmospheric cleansing also regenerate ozone and promote the formation of secondary aerosols, both of which act as potent air pollutants and short-lived climate forcers. Thus, the processes that maintain atmospheric self-cleansing capacity are intrinsically double-edged, sustaining a delicate balance highly sensitive to external perturbations.

AOC exhibits complex, nonlinear responses to anthropogenic emissions. For example, although increased NO_X concentrations generally enhance global OH production, thereby shortening the atmospheric lifetimes of CH_4 and CO, changes in NO_X emissions do not consistently lead to proportional changes in atmospheric concentrations due to OH-mediated feedbacks (Box 1 scenario c and d)[17]. In contrast, reductions in CH_4 emissions directly lower its atmospheric burden and decrease OH consumption, resulting in higher OH concentrations

(Box 1 scenario a). The behavior of CO is more complex. Although it dominates global OH turnover, its atmospheric abundance arises not only from direct emission but largely from CH_4 oxidation through HCHO intermediates. The latter process consumes about 2.5 OH radicals per CH_4 molecule, compared to only one OH per directly emitted CO molecule. As a result, CH_4 emissions exert a stronger influence on the atmospheric oxidation system, owing to their greater cumulative OH consumption and their role as the primary source of CO, whereas CO emission perturbations have a comparatively minor effect (Box 1 scenario b)[18]. Stabilizing AOC therefore necessitates carefully balanced, multi-pollutant reduction strategies (Box 1 scenario f).

Moreover, climate change may perturb AOC through multiple pathways. Elevated temperatures and water vapor levels could enhance OH production by accelerating ozone photolysis. However, this can be counteracted by the temperature-dependent acceleration of CH_4 oxidation kinetics, which consumes OH and may moderate the net increase in OH concentration (Box 1 scenario g and h). Furthermore, climate-driven increases in biogenic emissions, notably N_2O, contribute to stratospheric O_3 depletion. The resulting increase in tropospheric ultraviolet radiation elevates surface OH concentrations and shortens the lifetime of CH_4[19] (Box 1 scenario e). These interacting feedbacks complicate atmospheric predictions and highlight the necessity of integrated chemistry-climate assessments.

Quantifying AOC: Metrics and Interpretation

Quantifying AOC across scales remains a fundamental challenge, yet is essential for understanding atmospheric chemistry and climate interactions. Accurate quantification requires the careful selection of appropriate metrics aligned with specific research objectives. From a kinetic perspective, we distinguish two principal metrics: (1) the OH concentration, an intensive

property governing oxidative potential, and (2) the net O_3 production rate ($P(O_3)$), an extensive property characterizing chemical transformation rates. These complementary metrics serve distinct but interrelated roles in atmospheric process analysis.

OH concentration provides a foundational measure of AOC, particularly on larger scales. Atmospheric trace gases degradation is primarily driven by three key oxidants, namely OH, O_3, and NO_3. OH and NO_3 radicals exhibit extremely short lifetimes, typically under one minute, resulting in pronounced spatial heterogeneity tied to the distribution of their precursors. Appropriate spatiotemporal averaging of these radical concentrations yields a robust intensive parameter that quantitatively represents AOC while inherently accounting for variability. In practice, OH concentration is widely adopted as a proxy due to its central role in oxidizing most trace gases. Although this simplification may lead to a modest underestimation of total AOC, it provides a standardized and practical framework for cross-scale comparison. For long-lived species such as CH_4, decadal-average OH concentrations provide a robust measure of the atmosphere's self-cleansing capacity and remain the benchmark for global AOC assessment.

In the context of regional air quality, metrics of prompt oxidation rate, such as $P(O_3)$, offer more direct insight into secondary pollutant formation. This process involves the radical-driven oxidation of primary pollutants such as NO_X, VOCs, generating intermediate species whose conversion efficiency to secondary products like O_3 and SOA depends strongly on local chemical conditions. Tropospheric O_3 concentrations have approximately doubled since the pre-industrial era[23], suggesting a rise in $P(O_3)$ and thus an increase in AOC. In contrast, Earth system models indicate that global mean OH concentrations have varied by less than 10% over the same period[24], though uncertainties in

chemical mechanisms complicate these estimates[25]. This divergence underscores the need to tailor AOC definitions to the process and timescale of interest. The historical rise in O_3 reflects a coupled atmospheric response, where increased O_3 production from anthropogenic emissions accompanies OH stabilization through NO_X-mediated HO_X cycling, illustrating a nonlinear feedback within the oxidation system. Thus, $P(O_3)$ serves as a valuable diagnostic of chemical efficiency in pollutant formation, revealing aspects of oxidation chemistry that are not captured by OH concentrations alone.

In polluted environments with high aerosol loading, AOC assessment requires additional considerations. Multiphase chemistry necessitates supplementary metrics, such as O_X ($=O_3+NO_2$) and secondary aerosol mass fractions, to fully capture oxidation processes. Nevertheless, in this perspective, we focus mainly on gas-phase reactions to reduce system complexity, while clarifying how OH-mediated hydrocarbon oxidation couples air pollution with climate change.

Key Drivers and Chemical Regimes of AOC

The spatial distribution of AOC is primarily governed by emission patterns, with NO_X serving as the dominant driver of its variability[26]. This regulatory role manifests as a clear contrast between chemical regimes, where polluted urban regions are often NO_X-saturated, suppressing oxidation efficiency, whereas remote regions are typically NO_X-limited, with oxidative capacity tightly coupled to NO_X availability[20-21]. State-of-the-art box model simulations capture this contrast, revealing the distinct responses of OH concentration and in-situ $P(O_3)$ to varying NO levels across urban, forest, and marine environments (Figure 2)[27]. A key feature emerging from these simulations is the divergent behavior of the two AOC metrics across NO concentrations spanning six orders of magnitude. OH concentrations remain relatively stable, maintaining levels between (1-5)×

10^6 cm^{-3} with less than fivefold variation. In contrast, $P(O_3)$ exhibits pronounced nonlinearity, transitioning from slightly negative values in pristine marine air (ppt NO levels, mixing ratios of parts per trillion) through maximum production rates (tens of ppb/h, parts per billion per hour) in moderately polluted urban air (a few ppb NO, parts per billion) to nearly zero in heavily polluted urban air (tens of ppb NO).

Furthermore, while OH concentrations at a given NO level can differ across regimes (Figure 2a), $P(O_3)$ displays a smooth, consistent functional relationship with NO that is largely independent of the local environment (Figure 2b). This divergence arises from regime-specific VOC characteristics, particularly their abundance and chemical complexity, which modulate the conversion efficiency of OH radicals to peroxy radicals. Globally, a positive feedback links O_3 and OH, where VOCs oxidation drives O_3 production, which in turn photolyzes to generate OH and accelerates further oxidation. In the marine boundary layer (MBL), however, limited OH reactants (mainly CH_4 and CO) suppress radical propagation, while extremely low NO concentrations restrict O_3 production, constraining the sustainability of AOC. This establishes a fundamental spatial decoupling: although the global cleansing capacity for long-lived gases like CH_4 is determined by moderate OH over the vast MBL, the chemical potential for O_3 production is concentrated over continents, particularly in urban and suburban areas. The hemispheric-scale transport of O_3, which has an atmospheric lifetime of approximately one month, critically couples these chemically distinct domains, enabling OH production via O_3 photolysis even in remote areas where local O_3 chemistry results in net loss (e.g., through reactions with OH, HO_2 and halogen species in the MBL)[28]. This O_3-mediated chemical buffering effectively extends AOC globally, facilitating pollutant degradation even in NO_X-limited environments.

Nevertheless, at smaller scales, the oxidation capacity is not unlimited and can be exceeded under high emission loads, particularly in urban areas[29]. When local AOC recovery mechanisms are overwhelmed, excess pollutants are transported downwind, moving across chemical regimes and undergoing oxidation over broader spatial scales. This redistribution mechanism, which links local chemistry to global AOC, represents a critical feature of atmospheric self-cleansing. It underscores the necessity of multi-scale emission management strategies that explicitly account for the inherent spatial heterogeneity and nonlinearity of atmospheric oxidation processes.

DIVERGENT AOC TRENDS AND REGIONAL POLICY LESSONS

A central question in AOC research involves determining long-term trends in OH concentrations, given their critical role in regulating the removal of CH_4 and its associated climate impacts. Reliable quantification of OH variability is, therefore, essential for understanding how anthropogenic emissions influence the atmospheric lifetime and burden of CH_4. The complex variations in the decadal-scale growth rate of atmospheric CH_4 underscore the urgency and importance of this quantitative effort.

Observational records reveal that although CH_4 has generally exhibited a long-term increasing trend since industrialization, this trend was interrupted by a distinct pause between 2000 and 2006. While the long-term increase is largely attributed to the rise in anthropogenic emissions[30], the causes of the pause remain debated. Proposed explanations include an increase in OH concentrations enhancing CH_4 oxidation, a temporary halt in CH_4 emissions growth, or a combination of both[31-32]. Definitive attribution, however, is complicated by substantial uncertainties in quantifying key processes such as

OH concentration estimates, wetland emissions, fossil fuel leakage, and microbial sources. Specifically, each of these factors contributes approximately 10% uncertainty to the CH_4 budget. Cumulatively, these uncertainties exceed the observed CH_4 growth rate of ~0.5% yr^{-1}, making it difficult to confidently attribute both the historical pause and the subsequent renewed growth.

Model simulations from CESM2/WACCM6 initial-condition ensembles suggest a sustained increase in global OH after the year 2000 (Figure 3)[33], which may have partly attenuated CH_4 growth between 2000 and 2006, though its magnitude appears insufficient to fully account for the stabilization. The subsequent leveling-off of OH concentrations after 2007 coincides with the resumption of CH_4 growth[34], reinforcing the coupling between OH variability and CH_4 trends. The unprecedented acceleration in CH_4 growth during the 2020 COVID-19 lockdowns further illustrates how abrupt reductions in NO_X emissions can perturb global OH levels, leading to a substantial increases in CH_4 growth[4], although elevated wetland emissions also contributed significantly to the anomaly[35-36]. While this Perspective focuses on elucidating the linkages between global OH concentrations and regional air pollution rather than attributing specific CH_4 trends, it underscores the necessity for Earth system models to adequately resolve these critical chemistry-climate interactions.

As global OH concentrations are primarily governed by NO_X emissions and ultraviolet radiation[37], a realistic representation of these drivers is critical for projecting future OH levels and adjustments in CH_4 atmospheric lifetime. Among these drivers, natural NO_X sources, particularly lightning, play a critical yet poorly constrained role in the global oxidative budget[52]. These emissions are strongly modulated by weather and climate, and their representation in models remains a major source of uncertainty. Emerging evidence indicates

that lightning directly generates OH radicals alongside NO, with OH production efficiencies ranging from 2% to 16%[53]. As a key driver of wildfires and with frequency projected to increase 41% by the 2090s under RCP6.0 climate scenario[54], lightning constitutes one of the largest uncertainties in projecting future OH levels and AOC[51]. The uncertainties introduced by these complex and variable drivers make robust observational constraints on OH levels especially critical for evaluating model simulations. However, observational constraints on OH beyond 2014 remain subject to significant uncertainty, primarily due to limitations of the methyl chloroform (CH_3CCl_3) tracer method, whose accuracy has been severely compromised as concentrations decline below 5 pptv[50]. This scarcity of robust observational data after 2014 critically limits our ability to evaluate model simulations and refine future projections.

The regional distribution of OH concentrations provide additional critical insights into the atmospheric lifetime and burden of CH_4. As outlined in Section 2, the spatial decoupling between global OH and O_3 as AOC metrics reflects a fundamental heterogeneity in oxidation intensity. This heterogeneity manifests through three key mechanisms: First, tropospheric O_3 production is concentrated primarily in the Northern Hemisphere mid-latitudes[51], where major urban and industrial regions (e.g., China, the United States, and Europe) are located. NO_X emissions from these areas not only drive intense local photochemistry but also help sustain OH levels in remote forested and marine environments, thereby facilitating the global removal of CH_4 (Figure 2). Second, the spatial heterogeneity of OH concentrations leads to a pronounced tropical dominance in the oxidation of VOCs and, consequently, in atmospheric CH_4 consumption[24]. Interhemispheric OH differences further modulate CH_4 distribution and modify the climate-chemistry interactions in response to anthropogenic emissions[38]. Third, and crucially for policy, regional-scale OH

variations serve as a key metric for evaluating the effectiveness of air pollution mitigation strategies, which are typically implemented at national or subnational scales.

The contrasting trends between China and the United States offer a clear illustration of this principle (Figure 3). In the United States, OH concentrations have declined alongside improvements in air quality[39]. In China, by contrast, OH has increased consistent with observed rise in O_3[40] and secondary aerosols[41], despite reductions in primary emissions following implementation of the national Air Quality Action plan in 2013[42]. This divergence can be partly attributed to differing NO_X reduction strategies. China's integrated approach targeting both NO_X and VOC control[43] may help sustain regional AOC while improving air quality[18], whereas the predominant focus on NO_X reduction in the United States[44-46] contributes to a more pronounced regional OH decline. Such regional-scale OH reductions, while beneficial for local air quality, may influence AOC beyond local scales[9] and introduce climate trade-offs by weakening the CH_4 sink, as evidenced during the COVID-19 lockdowns[4]. Current Earth system models, however, still do not fully capture these climate impacts resulting from air quality-driven changes in CH_4 loss rates, despite recent efforts to incorporate such chemistry-climate interactions[47-49].

At regional and urban scales, the intricate interplay between reactive nitrogen and carbon chemistry emerges as a key regulator of AOC. This regulation is particularly evident in urban environments, where primary radical sources such as nitrous acid (HONO) and HCHO photolysis often dominate over O_3 photolysis. This dominance explains the occurrence of winter photochemical smog in high-emission regions, such as areas with extensive natural gas production or petrochemical industries like the Utah Basin in the United States, where significant emissions of oxygenated volatile organic

compounds (OVOCs) drive winter O_3 pollution[56-57]. In China, unique topographic features favor large-scale pollution accumulation, especially in the North China Plain under stagnant atmospheric conditions, creating ideal environments for studying these processes. Radical budget analyses based on in situ measurements confirm the roles of reactive nitrogen species beyond NO_X, particularly HONO and nitryl chloride ($ClNO_2$), in the formation of O_3 and aerosols[58-59].

Beyond these established mechanisms, the understanding of AOC is further complicated by recently identified chemical processes whose impacts are not yet fully constrained[60-61]. For instance, regional fertilization via agricultural activities releases HONO and N_2O, which may promote O_3 pollution[62], offset pollution control measures[63], or alter the CH_4 lifetime through troposphere-stratosphere interactions[9]. Similarly, the growing recognition of reactive halogen chemistry reveals its role in urban air pollution and climate change[64-65], offering new insights for controlling secondary pollutants. However, the chemical behavior of short-lived halogen species remains inadequately characterized. Of particular interest are their indirect climatic effects mediated through changes in AOC, an aspect not yet incorporated into current Earth system models[66]. In conclusion, these non-conventional pathways significantly enhance local AOC under intensive anthropogenic emissions, accelerating the formation of secondary pollutants and posing novel challenges for air quality management. The resulting complexity underscores that the limited understanding of emission profiles and chemical mechanisms remains a major source of uncertainty in air quality prediction and regulation worldwide[67].

The formation of secondary pollution is further complicated by seasonal variations in AOC. Summer O_3 pollution is primarily driven by OH-initiated

oxidation pathways, whereas winter secondary aerosol formation involves both gas-phase and multiphase processes[68-69]. Resolving these dynamics requires long-term observational records of OH concentrations, which are critical for identifying the factors controlling AOC across timescales from diurnal to seasonal[70]. The importance of such datasets is underscored by a five-year in situ study, which revealed a robust correlation between OH concentrations and O_3 photolysis rates ($j(O^1D)$) that persisted across diverse chemical regimes[71]. Although geographically limited to a single rural site, these findings significantly advance our understanding of AOC drivers. Furthermore, decadal-scale OH measurements are essential for tracking the evolution of air pollution and its associated climate feedbacks. We therefore propose establishing a multi-scale observational network by employing comprehensive techniques across local, regional and global scales (see Box 2). Such an integrated network would greatly enhance our ability to interpret complex climate-chemistry interactions through long-term, high-quality data.

AN INTEGRATED AOC GOVERNANCE FRAMEWORK

For future applied and scientific investigations, we propose exploring the role and fate of AOC through a consolidates framework that unifies the scientific and technical basis to address both air pollution and climate change, consistent with existing environmental conventions (Figure 4). Future atmospheric dynamics will be shaped by energy system transitions toward climate mitigation targets, alongside climate-driven shifts in natural emissions, including biogenic, wetland, lightning-derived sources. The nonlinear atmospheric response to emissions, mediated by AOC mechanisms, exhibits particular sensitivity to declining NO_X emissions, a trend expected to intensify. To optimize regional air quality strategies, mitigation roadmaps must rigorously account for AOC feedbacks, necessitating targeted research to strengthen the

scientific basis for implementing a win-win strategy that simultaneously improves air pollution control and climate change mitigation. These efforts must also be contextualized within the broader scope of other international environmental conventions.

Navigating the Energy Transitions Green Paradox

The global energy transition is accelerating decarbonization across fossil fuel-intensive sectors, including heavy industry, transportation, and power generation. Current technological pathways for emission reduction can be classified into three categories based on their impacts on atmospheric chemistry: (1) the adoption of zero-emission technologies, such as photovoltaics, wind, hydroelectric, and nuclear power, enables the simultaneous reduction of CO_2 and NO_X emissions; (2) the shift to hydrogen (H_2) fuel cells reduces CO_2 and NO_X emissions at the point of use but can be accompanied by unintended release of reduced chemical species, such as fugitive H_2 emissions across the supply chain; and (3) the combustion of H_2 or ammonia, as a replacement for fossil fuels, reduces CO_2 emissions but may be offset by elevated NO_X formation from high combustion temperature and by H_2 leakage.

The rapid adoption of electric vehicles (EVs) serves as a prime example of the potential atmospheric impacts of the zero-emission technological pathway in the energy transition. Shifting to EVs powered by grid electricity from renewable sources eliminates a primary source of CO_2 and especially NO_X emissions in urban environments, but this transition introduces complex side effects that require thorough investigation. The COVID-19 lockdowns provide a revealing natural experiment analogous in many ways to the EVs transition, as transportation emissions via internal combustion engine vehicles was drastically reduced during this time. This emission reduction revealed the nonlinear behavior and critical importance of AOC in secondary pollution

formation[72]. Global observations showed that although NO_X reductions improved air quality in some regions[73], China experienced secondary pollution bursts (O_3, particulate nitrate, and organics)[74-75] and global CH_4 loss rates declined undesirably[4]. These divergent, region-specific outcomes demonstrate that the impacts of fossil fuels phase-out depend strongly on local chemical regimes, shaped by the interplay of anthropogenic and biogenic emissions with meteorological conditions. Notably, the air quality benefits are localized, but the potential increases in CH_4 levels may exacerbate global climate impacts. This disparity creates disproportionate burdens for developing countries, underscoring a critical equity dilemma in climate mitigation policy. The EVs transition thus serves as a critical warning, highlighting the imperative to strategically manage AOC in order to navigate these inequities and complex trade-offs.

Given the anticipated near-term persistence of CH_4 and reactive carbon emissions, maintaining AOC will necessitate the strategic deployment of NO_X emissions, despite their inherent trade-off with local air quality. This approach is grounded in the spatial interdependence of the oxidation system: relocating NO_X-emitting industries to tropical regions, which are characterized by high oxidation capacity and function under NO_X-limited chemical regimes, could enhance the OH levels responsible for global CH_4 removal, while reducing ground-level O_3 and NO_X in populated areas[76]. This offers a promising pathway to co-managing air quality and climate mitigation. However, implementing such geoengineering strategies is not a direct decision but a complex prospect fraught with challenges. It first demands comprehensive evaluation across three critical dimensions: (1) the ethical assessment of pollution burden redistribution and its socioeconomic consequences, (2) a rigorous cost-benefit analysis comparing AOC gains against local environmental and health costs,

and (3) the establishment of equitable compensation frameworks, potentially integrated with carbon market mechanisms, to address regional disparities in environmental impacts. Furthermore, even if deemed ethically and economically viable, realizing this potential depends critically on the availability of accurate numerical models capable of simulating the complex interactions among meteorology, climate, and atmospheric chemistry in response to anthropogenic emission changes[55]. At present, the utility of Earth system models for this task is hindered by considerable uncertainties, including inaccuracies in emission inventories, oversimplified chemical mechanisms, and insufficient coupling of key climate-chemistry feedbacks. Therefore, advancing the predictive capability of these models is an essential prerequisite to translating the concept of emission redistribution into credible, policy-relevant strategies.

Hydrogen, as a clean energy carrier and carbon-neutral fuel, represents a promising alternative pathway toward carbon neutrality, especially as electrification generates surplus electricity for its production via electrolysis. However, the large-scale deployment of H_2-based technologies, whether in fuel cells or through combustion of H_2 or ammonia, introduces substantial atmospheric risks that extend well beyond engineering and economic constraints. The primary common risk is fugitive emission of H_2 and its precursor, CH_4, across the supply chain. For instance, the ‘blue hydrogen’ process, which utilizes CH_4 as feedstock to produce H_2 with CO_2 byproducts, offers one potential pathway to meet global energy needs. Yet even with perfect CO_2 capture, this approach remains prone to fugitive emissions of H_2 and CH_4 during transport and use, which could increase atmospheric CH_4 levels, thereby offsetting potential climate benefits[17, 77]. Additionally, H_2 combustion produces water vapor that may influence climate systems from

local to global scales[78], and could potentially impact stratospheric chemistry[79]. Furthermore, the use of ammonia (NH_3) as a potential H_2 carrier and fuel, may alter particulate acidity and exacerbate urban air pollution[80].

These emerging risks necessitate a full life-cycle environmental impact assessment of H_2 prior to large-scale deployment[81], particularly regarding the response of atmospheric self-cleansing capacity to shifting emission patterns, a relationship clearly demonstrated through historical OH records that reveal how energy system transitions have fundamentally reshaped AOC. Consequently, climate governance must undergo a fundamental paradigm shift by elevating AOC stabilization to the same strategic priority as CO_2 mitigation within policy frameworks. This is essential to avoid trading near-term gains for long-term crises, such as compromised AOC, accelerated CH_4-driven warming, and the inequitable pollution redistribution. This requires policies that integrate chemical feedback mechanisms, informed by advanced atmospheric research and historical evidence, to maintain Earth's self-cleansing capacity while achieving emission targets.

Advancing Next-Generation AOC Model Development

(a) Improving OH Chemistry in VOC-Rich Environments

Current atmospheric chemistry models systematically underestimate AOC in environments dominated by biogenic VOCs (BVOCs) and OVOCs, particularly under low-NO_X conditions[45,65,82]. This persistent model bias, evidenced by consistent discrepancies between measured and simulated OH radical concentrations[16, 83-84], points to critical gaps in our mechanistic understanding of VOCs oxidation pathways[26]. Key uncertainties involve the autoxidation mechanisms of BVOC- and OVOC-derived RO_2 radicals, their temperature-dependent H-shift tunneling kinetics, and the contribution of unimolecular RO_2 reactions to OH recycling[26, 85-86]. To address these gaps,

integrated laboratory, theoretical, and field studies are required to quantify the rates and products of RO_2 isomerization and fragmentation reactions under atmospherically relevant conditions. Incorporating these refined mechanisms into next-generation chemical models is essential for accurate projection of OH radical concentrations and AOC. This advancement is particularly urgent given ongoing policy-driven emission changes, such as afforestation increasing BVOC emissions and decarbonization reducing anthropogenic NO_X emissions, and will provide a more robust scientific foundation for climate and air quality policymaking.

(b) Enhancing Climate-Chemistry Feedback in AOC Projections

Biogeochemical feedbacks: Accurate projection of AOC under climate change necessitates an integrated understanding of biogeochemical feedbacks coupled with dynamic emission processes. Current models exhibit substantial uncertainties, particularly in representing climate-sensitive emissions and multi-scale chemical interactions. Addressing these gaps demands a systematic effort to enhance the mechanistic representation of key processes and to develop observationally constrained, predictive frameworks. A primary challenge lies in quantifying climate-driven emissions of reactive species. Rising temperatures amplify releases of VOCs from both anthropogenic volatile chemical products (VCPs) and BVOCs. Concurrently, natural methane emissions from wetlands and NO_X from soils and lightning are perturbed by warming, while increasing wildfire activity contributes substantial reactive gases and aerosols[87-90]. To reduce uncertainties in these fluxes, future work should prioritize the development of process-based emission modules responsive to climate forcing, supported by improved mechanistic parameterizations and multi-platform observational constraints. Advanced monitoring technologies, such as remote sensing, eddy covariance systems,

and chamber-based field measurements coupled with next-generation Earth system models, will be critical to capture temperature-dependent changes in AOC and to better project chemistry-climate feedbacks.

Tropospheric halogen chemistry: These complex interactions are further compounded by significant uncertainties in halogen chemistry[64-66], particularly within the MBL, where persistent knowledge gaps limit our ability to accurately quantify oxidative processes. Major challenges include insufficient kinetic data for critical reactions (e.g. IO+CH_3O_2), sparse measurements of reactive halogen species (RHS) such as IO and BrO that are essential for resolving their spatiotemporal distributions, and poorly constrained emission estimates that are likely to be intensified under climate change[66, 91]. An especially pressing priority is to refine the understanding of bromine (Br) in governing the atmospheric lifetime of mercury (Hg), which currently shows high variability (4 to 40 days) due to uncertainties in Br sources. Although Br has traditionally been ascribed to marine emissions, recent observations of elevated BrCl over the NCP in China suggest the presence of additional, poorly quantified terrestrial or anthropogenic sources[61]. Moreover, the activation pathways of RHS, primarily mediated by NO_X, introduce further complexity, as future reductions in NO_X emissions may fundamentally reshape these mechanisms in ways not yet captured by models. Addressing these gaps demands a targeted research strategy aimed at enhancing the identification of RHS source and refining their representation in model. Critical steps comprise expanding field measurements to better constrain spatiotemporal RHS variability, conducting laboratory studies to resolve kinetic parameters, and integrating refined halogen emission and chemistry modules into climate-chemistry models. Such advancements are essential for reducing uncertainties in projections of Hg cycling and enhancing the accuracy of halogen-mediated climate feedbacks.

Interactions with troposphere-stratosphere: Furthermore, evolving emission patterns due to intensified industrial pollution controls are elevating the relative importance of agricultural activities in atmospheric chemical processes. The increased use of fertilizer and expansion of agricultural activity are projected to enhance soil N_2O emissions, which may indirectly modulate atmospheric CH_4 levels via complex stratosphere-troposphere interactions[19, 92]. Stratospheric processes introduce additional complexity, with model ensembles suggesting that stratospheric ozone accounts for approximately 25% of surface ozone concentrations, although inter-model variability exceeds a factor of two[93]. Furthermore, emerging evidence indicates that stratospheric air intrusions can affect climate regimes by triggering large-scale new particle formation events[94], revealing previously underappreciated troposphere-stratosphere interactions that require systematic investigation under future climate scenarios. Efforts to constrain these linkages must integrate advanced observational networks with next-generation modelling frameworks. Critical pathways forward involve improving the mechanistic representation of agricultural N_2O and CH_4 coupling within Earth system models, achieving higher vertical resolution to quantify stratosphere-troposphere exchange processes, and deploying targeted field campaigns to validate the impact of stratospheric intrusions on particle formation and radiative forcing.

(c) Coupling Methane and Chemistry in Earth System Models

Current Earth system models predominantly rely on prescribed CH_4 concentrations rather than prognostic, emissions-based approaches. While emerging model frameworks are beginning to incorporate interactive methane cycling[47-49], many widely used models still fail to capture the nonlinear coupling between CH_4 emissions and atmospheric concentrations, neglecting critical

chemical feedbacks, particularly those involving radical chemistry. The prescribed-concentration approach implicitly parameterizes OH levels using simplified scaling factors for changes in OH precursors[18,95], introducing a fundamental inconsistency in the representation of atmospheric oxidation capacity. Significant uncertainties in CH_4 chemical loss rates, driven largely by poor constrains on global OH concentrations, further impede the adoption of emission-based approaches and amplify uncertainties in climate projections.

To advance Earth system modeling towards robust, emission-driven CH_4 simulations, a concerted effort is needed to reduce key mechanistic uncertainties and develop advanced model configurations with fully interactive CH_4 chemistry. A critical priority is to significantly reduce uncertainties in atmospheric chemical sink processes, particularly the representation of OH chemistry. This requires refining the spatiotemporal variability of OH concentrations within models by improving the mechanistic understanding of radical chemistry and the representation of key drivers, such as photolysis rates, NO_X emissions, and VOC interactions. Enhanced coupling between tropospheric chemistry and climate dynamics is also essential to capture feedback mechanisms that modulate CH_4 lifetime under evolving scenarios. Concurrently, process-based representations of methane sources, especially climate-sensitive wetlands, thawing permafrost, and inland waters, require improved mechanistic modelling of their hydrological, ecological, and biogeochemical interactions. These modelling efforts must be strongly supported and constrained by multi-platform observations that integrate top-down remote sensing with bottom-up field measurements, enabling improved emissions quantification and model validation through advanced data assimilation. Ultimately, integrating these components into coupled emission-chemistry modules that dynamically link anthropogenic and natural

sources with atmospheric oxidation processes will be crucial. Such advancements will support a robust transition from concentration-driven to emission-driven CH_4 representation, enabling more reliable projections of AOC and better-informed evaluation of climate-air quality policy interactions.

Global AOC Monitoring and Science-Based Policy

The transition to low-carbon energy systems introduces complex atmospheric trade-offs, underscoring the need to systematically monitor AOC as a critical metric of Earth's atmospheric resilience. A robust global AOC observation network would provide the essential data required to evaluate OH concentrations, which govern the atmospheric self-cleansing capacity and the lifetime of CH_4, as well as total OH reactivity that serves as a key proxy for radical loss processes in atmospheric models. As outlined in Box 2, such an integrated monitoring system would deliver quantitative, policy-relevant metrics across spatial and temporal scales, bridging fundamental science and decision-making.

Implementation this network requires coordinated advances across multiple measurement technologies (Box 2). Existing ground-based atmospheric supersites require upgrades with next-generation instrumentation capable of simultaneous radical measurements and reactivity quantification. The high costs of these technologies must be reduced through innovation in laser spectroscopy and sensor miniaturization to enhance accessible. Concurrently, novel satellite remote sensing methodologies must be developed to achieve global mapping of OH distributions, extending coverage beyond ground-based limitations[96]. These parallel developments require sustained international funding to establish standardized measurement protocols, ensure cross-platform data compatibility, and maintain the long-term monitoring necessary for detecting critical climate-chemistry feedback mechanisms.

The proposed AOC monitoring network offers equally significant scientific and policy applications. By characterizing spatiotemporal patterns in AOC, it would enable optimization of emission reduction strategies. For instance, guiding industrial sitting decisions that balance local air quality goals with global atmospheric cleansing needs. The network's data products would provide empirical benchmarks for assessing compliance with international climate agreements like the Global Methane Pledge and Paris Agreement. Furthermore, integration of AOC metrics into Earth system models would help resolve current uncertainties in CH_4-OH feedback mechanisms that currently constrain climate projections.

To operationalize this framework, we propose establishing an International AOC Science Alliance that brings together atmospheric chemists, remote sensing experts, and policy specialists. This consortium would oversee three primary functions: (1) harmonization of measurement standards and quality assurance across observational platforms; (2) development of open-access data repositories featuring real-time analytics capabilities; and (3) translation of scientific observations into actionable policy guidance for clean energy transitions. Through such a coordinated global effort, it becomes feasible to navigate the complex atmospheric impacts of decarbonization while preserving the Earth's self-cleansing capacity during this critical transition.

GOVERNING AOC FOR CLIMATE AND AIR QUALITY CO-BENEFITS

The transition toward carbon neutrality confronts a critical atmospheric governance challenge: sustaining Earth's self-cleansing capacity while simultaneously improving regional air quality and meeting global climate objectives. Policymakers urgently need predictive, systemic tools capable of

quantifying the trade-offs and co-benefits between air quality management and climate mitigation. However, a significant gap persists between current atmospheric science capabilities and policy requirements. Existing climate governance frameworks, including the Paris Agreement and Global Methane Pledge, often lack mechanistic links to atmospheric chemistry, leaving regulators without actionable insights into how emission changes impact oxidative capacity, pollutant lifetimes, or secondary formation. To bridge this gap, we propose an innovative AOC framework that translates complex chemical processes into policy-operational metrics. This approach moves beyond reactive regulation by embedding atmospheric feedbacks directly into policy design, enabling proactive and integrated decision-making. We call for the adoption of dynamic, chemistry-informed governance tools that align emission pathways with AOC stability, ensuring that climate strategies do not inadvertently compromise atmospheric resilience. Furthermore, we urge enhanced collaboration between scientific and policy communities to co-develop scalable, regionally tailored approaches that maintain oxidation capacity throughout deep decarbonization. Looking forward, AOC must be elevated from a passive chemical metric to a central governance pillar, enabling smarter and more adaptive climate policy grounded in the realities of atmospheric science.

Acknowledgements:

This work was supported by the National Natural Science Foundation of China (grant 22325601, 42475110, and 22406003), the National Key R&D Program of MoST China (2023YFC3706100, 2024YFC3713601, 2024YFC3712902, and 2023YFC3710900).

Author contributions:

Z.F.T. and X.F.M. designed and performed the study, and drafted and revised

the manuscript. K.D.L. and Y.H.Z. supervised the project and reviewed and revised the manuscript. X.L., Q.D.Z., and F.R. created the figures and provided analytical comments. A.N., H.F., A.W., L.W., D.H., S.B., and X.Z. contributed to data interpretation, manuscript discussion, and critical revision.

Declaration of interests:

The authors declare no competing interests.

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Figures and Tables:

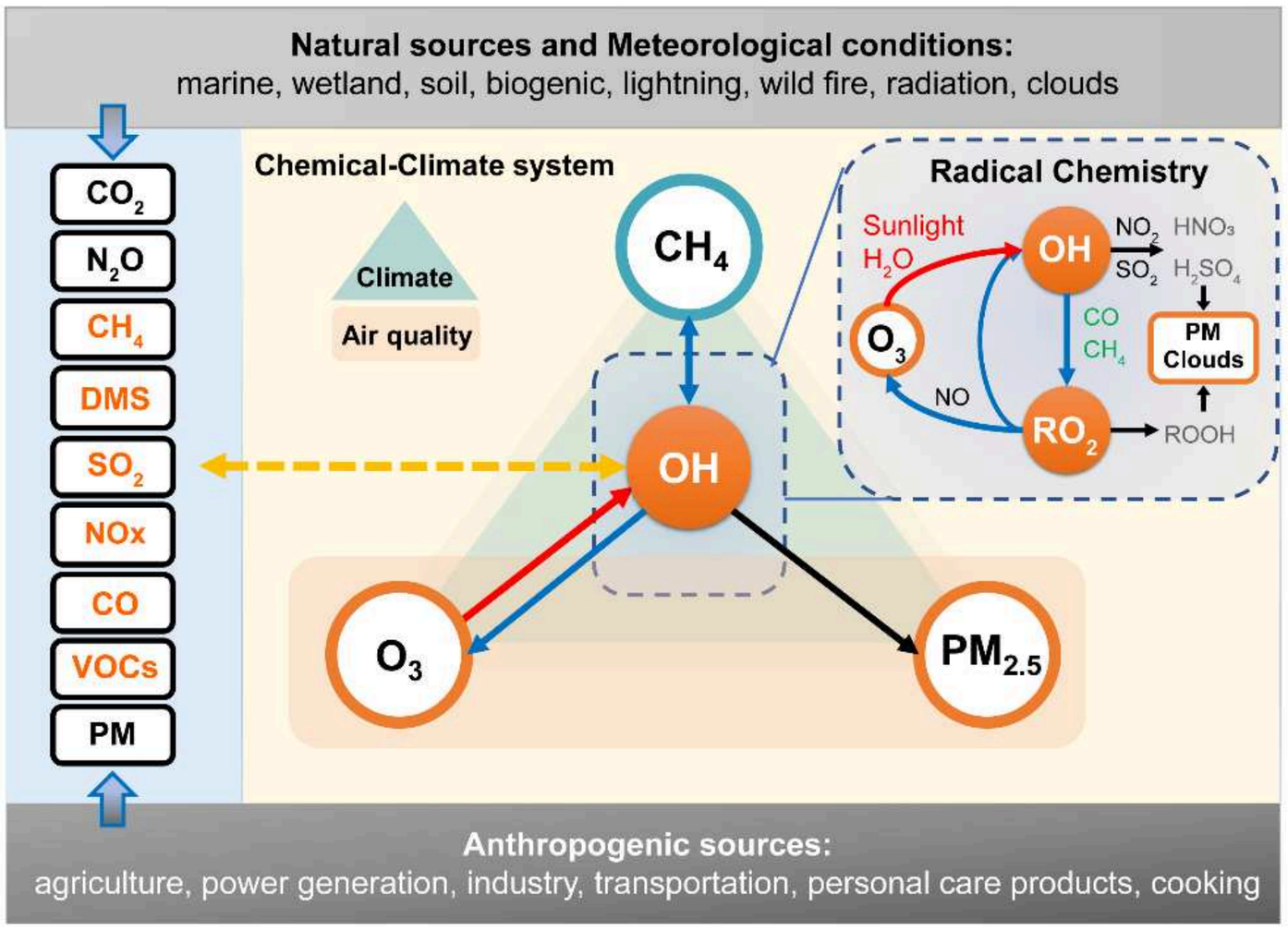

Figure 1: Schematic representation of tropospheric chemistry-climate interactions. OH governs the atmospheric lifetime of CH_4, a potent greenhouse gas, and drives the formation of secondary pollutants such as ozone and $PM_{2.5}$, which also act as significant climate forcers. It plays a critical role in linking air pollution and climate forcing by mediating key atmospheric chemical processes. Orange-highlighted species in the left panel represent reactive gases from anthropogenic and biogenic sources that undergo OH-initiated oxidation, while those marked in black denote emitted non-reactive greenhouse gases and pollutants. Blue arrows indicate the oxidation pathways of reactive carbon compounds (e.g., CH_4, CO, and VOCs), leading to the formation of secondary pollutants including O_3 and $PM_{2.5}$. The red arrow denotes the photolysis of O_3 to $O(^1D)$, followed by reaction with water vapor, representing the dominant global source of OH. The black arrow denotes the oxidation processes involving NO_X, SO_2, and VOCs that contribute to the production of secondary aerosols, such as nitrate, sulfate, and organic aerosols. The inset presents a simplified scheme of global tropospheric photochemistry. CH_4 serves as a key representative of OH-oxidized gases (including CO, H_2, VOCs), producing HO_2 and RO_2; RO_2 is a placeholder for peroxy radicals like HO_2 and RO_2 which react with NO to form NO_2; ROOH represents peroxides (e.g., HOOH, ROOH, and ROOR), which act as critical precursors to particulate matter (PM) and influence cloud formation, thereby affecting climate.

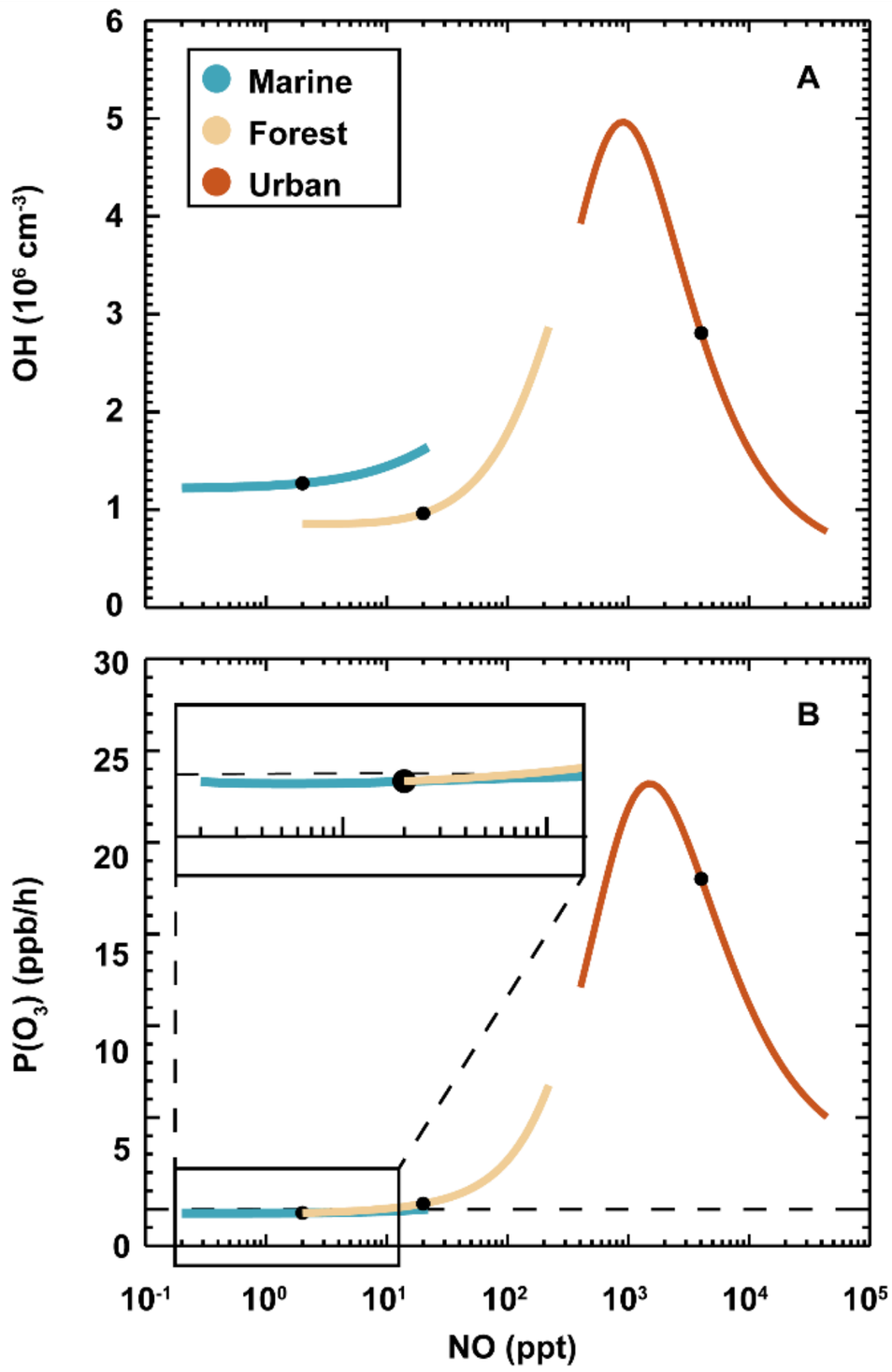

Figure 2: Dependence of OH concentration and net ozone production rate ($P(O_3)$) on NO levels across marine, forest, and urban environments. Simulations were conducted using a chemical box model incorporating the RACM2-LIM1 mechanism, which has been validated against field campaigns[27]. The principal distinction among the simulated environments lies in their VOC compositions. VOC profile were derived from field observations at representative locations: the tropical Atlantic Ocean (marine)[28], Amazon Forest (forest)[97], and Beijing (urban)[98]. In each scenario, VOC concentrations were held fixed at observed levels, as were other trace gases and meteorological parameters. Simulations were conducted under clear-sky, summer-like conditions using daytime-averaged values (08:00-17:00 local time) and run for one week to achieve steady state. To explicitly evaluate the NO dependence, the observed NO concentration at each site was systematically scaled from 0.01 to 100 times the site-specific mean value (denoted by black dots). $P(O_3)$ represents the net ozone production rate, calculated as the difference between gross ozone formation from NO oxidation by peroxy radicals and total oxidant loss via ozone reactions with radicals and NO_2. The detailed methodology for this calculation has been described previously[99].

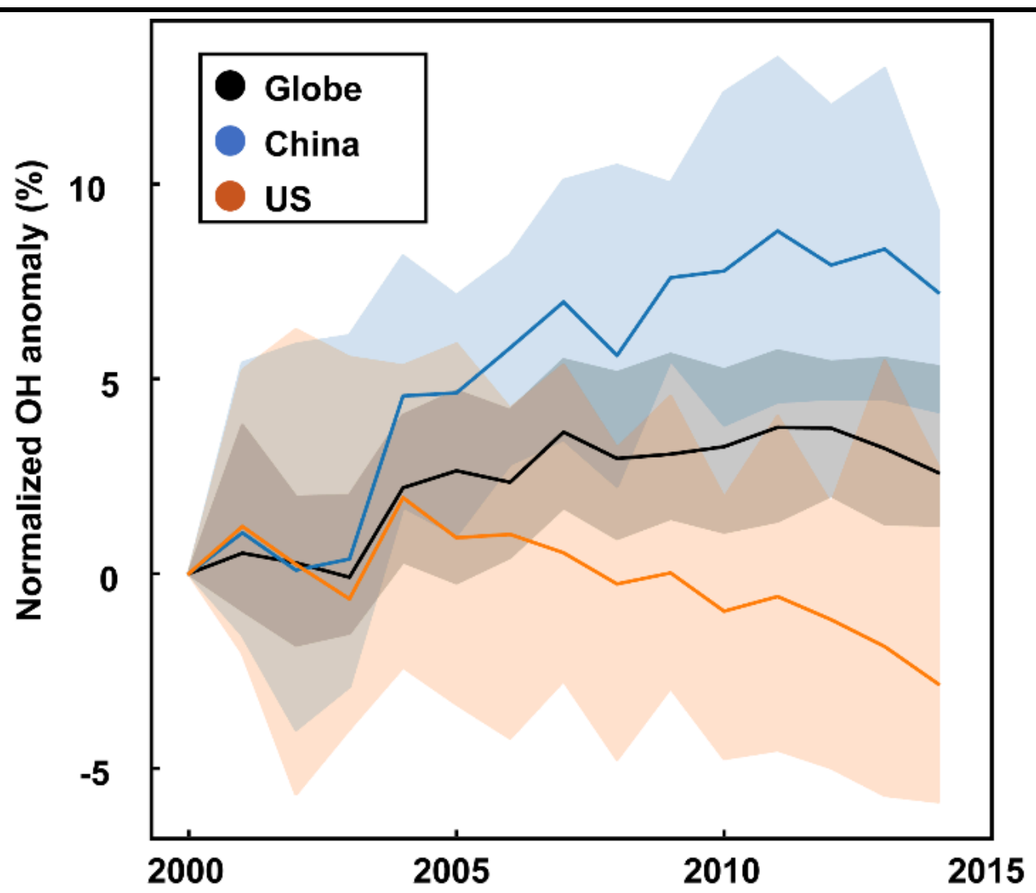

Figure 3: Time series of annual mean OH concentrations for China, the United States, and the global average, normalized to the year 2000. OH concentrations are derived from a 15-member initial-condition ensemble simulation conducted with the Community Earth System Model version 2-Whole Atmosphere Community Climate Model version 6 (CESM2-WACCM6). All simulations used identical boundary conditions, with variations across members resulting from perturbations in initial conditions[33]. The model covers the period 2000 to 2014 at a spatial resolution is 0.96° × 12.5°. The shaded regions represent the range between the minimum and maximum values across the ensemble members.

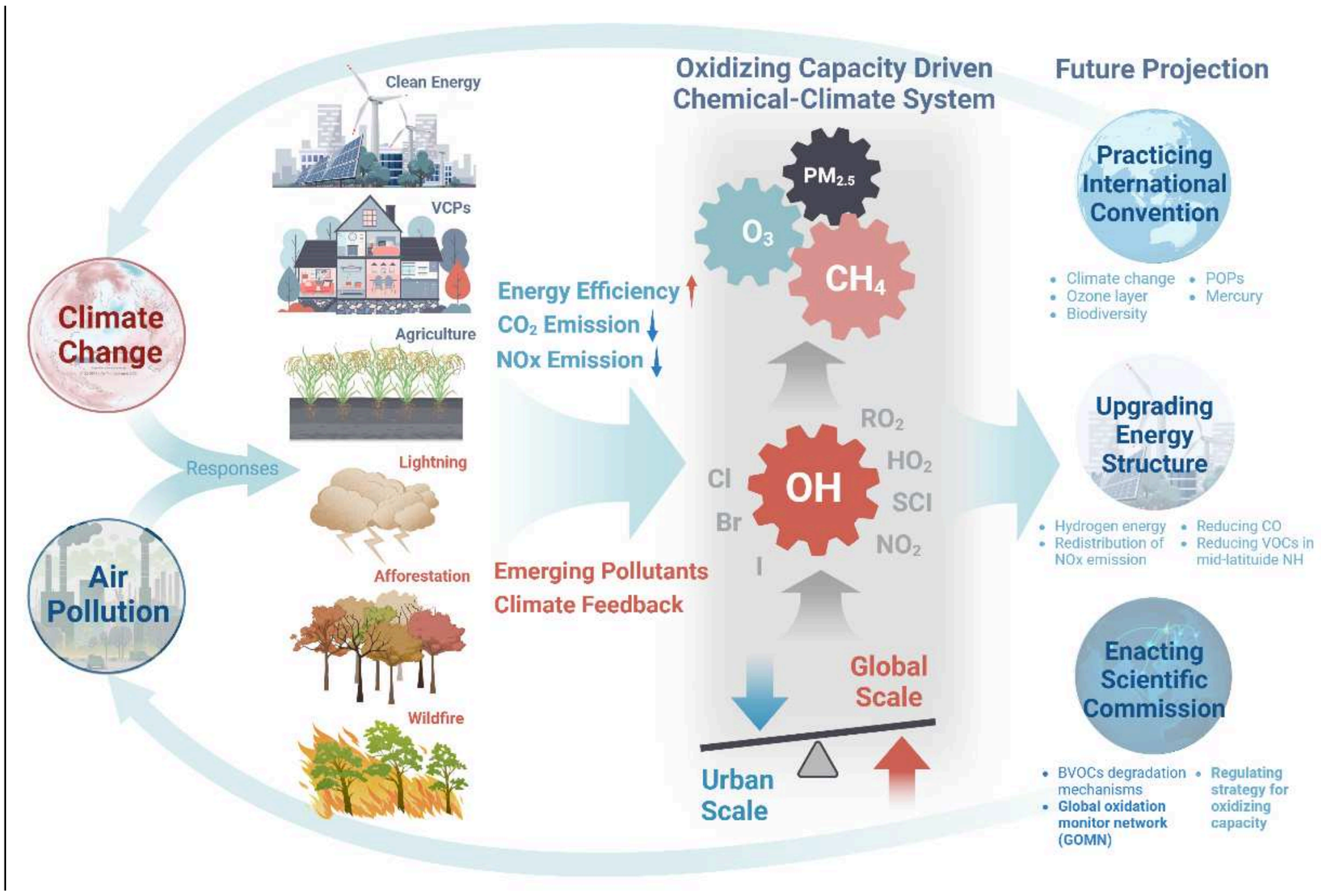

Figure 4: A proposed framework for future decision-making from the perspective of AOC. The transition to carbon neutrality will substantially alter

energy production and associated emission patterns. Simultaneously, natural emissions, such as those from biogenic sources, wetlands, and lightning, respond to ongoing climate change. The AOC critically influences the atmospheric lifetime of trace gases and the conversion of pollutants. To support effective regional air quality improvement, a scientifically grounded mitigation strategy should seek to regulate the AOC through balanced regional and global distribution.

Box 1: Global atmospheric composition responses to emission or boundary condition perturbations.

Box 1 summarizes the projected response of global concentrations of CH_4, CO, O_3, OH, and NO_X concentrations to minor perturbations in emissions or boundary conditions. An upward arrow (↑) indicates a positive response, a downward arrow (↓) a negative response, and a dash (-) no significant change. In scenario a, reduced CH_4 emissions decrease atmospheric CH_4 levels and OH consumption, leading to higher OH levels. In scenario b, although CO dominates global OH turnover, CH_4 emissions exert a stronger influence on atmospheric oxidation system because of their greater cumulative OH consumption (2.5 vs. 1 OH radical per molecule) and their role as the primary source of CO, making CO emission perturbations comparatively minor[24]. In scenario c and d, higher NO_X concentrations generally enhance OH production on a global scale, thereby shortening the atmospheric lifetimes of CH_4 and CO and reducing their concentrations. However, atmospheric NO_X levels do not respond proportionally to emission changes, and O_3 formation becomes relatively insensitive to NO_X due to OH-mediated feedbacks. Scenario f involves simultaneous reduction of CH_4, CO, and NO_X emissions by the same factor. Scenario e and g represent enhanced ultraviolet radiation and elevated water vapor, respectively, both promoting OH production via ozone photolysis. In scenario h, elevated temperature perturbs atmospheric chemistry by accelerating CH_4 oxidation (an OH-consuming process that reduces it concentration), while also promoting OH production via ozone photolysis.

Initial change		**Responses**					
Scenario	Emission changes	CH_4	CH_4 lifetime	CO	O_3	OH	NO_X
a	↓ CH_4	↓	↓	↓	↓	↑	↓
b	↓ CO	-	-	↓	-	-	-

c	↑ NO_X	↓	↓	↓	-	↑	-
d	↓ NO_X	↑	↑	↑	-	↓	-
e	↑ Sunlight	↓	↓	↓	↓	↑	↓
f	↓ CH_4, CO, NO_X	↓	-	↓	↓	-	↓
g	↑ H_2O	↓	↓	↓	↓	↑	↓
h	↑ Temperature	↓	↓	-	-	-	-

Box 2: Summary of measurement techniques and models for interpreting AOC for different scales.

On a global scale, the averaged OH concentration could be derived from the decay of the CH_3CCl_3, whose lifetime is similar to that of CH_4 and thus an ideal substance to reflects the chemical sink of CH_4. The production and emission of CH_3CCl_3 has ceased by early 2000s providing a clear decay trend to calculate the OH concentration for the last decade. However, the CH_3CCl_3 concentrations has recently dropped to below 5 pptv, substantially reducing the accuracy of OH estimates. It is therefore urgent and critical to identify an alternative to CH_3CCl_3. An ideal alternative may be either a natural atmospheric component or an intentionally introduced compound, but it must satisfy the following criteria: well-defined sources, exclusive reactivity with OH radicals, an atmospheric chemical lifetime comparable to methane, and minimal adverse effects on human health or ecosystem. ^{14}CO is produced in the stratosphere from the interaction of cosmic rays with nitrogen (^{14}N). This ^{14}CO then diffuses into the troposphere and reaction with OH which is a major sink for CO in the atmosphere. As the lifetime of CO is only about a month, this method could provide hemispheric and continental OH concentrations with a time resolution of months. For regional and local scale, the metric of interest for AOC quantification shifts to the turnover rates of oxidations. This requires both the OH concentration and reactivity. Direct OH measurement can be achieved by laser-induced fluoresce or chemical ionization mass spectroscopy[100]. The chemical reactivity of the airmass could be obtained by total OH reactivity measurements. These instrumentations could be equipped on ground-based or airborne platforms to provide local or regional AOC information. Here shows the summary of measurements techniques and models for interpreting AOC for different scales.

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Resolution Scale	Metric of interest	Requirement of measurements	Model	Possible improvement
Global	Decadal averaged [OH] for determining CH_4 lifetime and background O_3 production	CH_3CCl_3	Earth system model	New tracers (natural or artificially added)
Continental	[OH] variation in the time scale of months	^{14}CO	Global model	Interactions between air pollution and climate change
Regional	Regional [OH]	Spatial-averaged measurement of regional OH concentration and total reactivity	Regional model	Vertical profile of oxidizing capacity, relation to secondary pollution
Local	[OH] and total OH reactivity to test against current chemical mechanism	In situ OH, total and speciated reactivity	zero-dimension model	Atmospheric flagship stations (comprehensive measurements of other oxidants and intermediates species, covering representative stations of forest, marine, urban), BVOCs degradation mechanisms, OH parameterization

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