

A Study on Bridging Chemical Transformation and Climate Feedback in the Earth System

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Abstract

The atmosphere operates as a vast and complex chemical reactor, where minute-scale transformations exert profound influence on planetary-scale climate stability. This research investigates the multi-scale coupling between reactive tropospheric chemistry and large-scale climate feedbacks, challenging traditional modeling approaches that often divorce chemical kinetics from dynamic processes. By integrating high-resolution chemical transport models (CTMs) with comprehensive Earth System Models (ESMs), we map the flow of energy and matter from the molecular initiation of photochemistry to global radiative forcing. We demonstrate that the sensitivity of the tropospheric oxidant budget—driven by the concentration of hydroxyl radical (OH) and ozone—is critically modulated by evolving biogenic volatile organic compound (BVOC) emissions and aerosol composition. Crucially, the study identifies a dominant positive feedback loop: regional warming enhances BVOC emissions, which increases secondary organic aerosol (SOA) nucleation, leading to significant alterations in cloud condensation nuclei (CCN) population and subsequent cloud albedo. While this chemical-aerosol pathway exerts a net cooling tendency locally, the concurrent chemical lifetime reduction of potent greenhouse gases (GHGs) like methane introduces a competing warming effect at the global scale. This analysis underscores the inherent fragility of the atmospheric system and emphasizes that accurate climate projections fundamentally rely on resolving the non-linear coupling processes that bridge the micro-scale crucible of reactivity with macro-scale climate dynamics. Here we present the review of the system.

Keywords: Chemical Transformation, Climate Feedbacks, Earth System, BVOC, CCN,

INTRODUCTION

The atmosphere is often envisioned as a vast, passive envelope of gas—a simple blanket of nitrogen, oxygen, and trace warming agents that envelops the planet. In reality, it is the planet's most dynamic and complex chemical reactor, a system where events occurring on the scale of a nanometer and a millisecond ultimately govern the global energy balance over centuries.

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Understanding Earth's climate is fundamentally an exercise in navigating vast disparities in scale. Atmospheric processes form a continuous, interdependent chain, from the transient life of an oxidative radical to the multi-decadal circulation patterns that define the global climate. This journey, from the smallest chemical transformation to the largest climate feedback, is the essence of modern atmospheric science [1,2].

The Micro-Scale Crucible: Chemical Transformation

At the molecular scale, the atmosphere is a

relentless machine governed by photochemistry and radical reactions that are constantly occurring. This is the "engine room" of atmospheric processing, operating on timescales of seconds to hours.

The central player in this molecular drama is the hydroxyl radical (OH). Formed primarily when UV light interacts with ozone and water vapor, the OH radical is intensely reactive and ubiquitous, earning it the moniker "the detergent of the troposphere."

The primary roles of OH are transformation and removal. It scrubs the air, initiating the breakdown of virtually every gas emitted by human activities or natural sources. This is critical for climate science because chemical transformations dictate the atmospheric lifetime of greenhouse gases.

Consider Methane (CH₄). Methane is a potent warming agent, but unlike CO₂, which is removed primarily by oceans and the biosphere, CH₄ is removed predominantly by its reaction with OH. Therefore, the concentration of OH dictates the duration of methane retention in the atmosphere before being oxidized to less potent compounds (such as CO₂ and H₂O) [3,4].

This illustrates the first fundamental link: a tiny, short-lived chemical entity (OH) holds the global lever controlling the lifetime and, therefore, the warming potential of a long-lived greenhouse gas (CH₄).

Bridging the Gap: From Gas to Particle

When gaseous pollutants (such as sulfur dioxide or volatile organic compounds) are oxidized by OH, they transform into less volatile, heavier molecules (e.g., sulfuric acid and oxidized organic compounds). These oxidized molecules are prone to aggregation through a process known as nucleation.

Aerosols, which are tiny liquid or solid particles suspended in air, are the physical manifestation of chemical transformation. They bridge the gap between chemistry and dynamics by rapidly growing from nanometer-sized clusters to micrometer-sized atmospheric players [5]. These particles are crucial because they fulfill two major roles that profoundly affect the climate.

- *Radiative forcing:* Aerosols directly scatter incoming solar radiation back into space, providing a net cooling effect on the planet (a process known as the direct aerosol effect).
- *Cloud formation:* Aerosols act as Cloud Condensation Nuclei (CCN). Without a surface to condense on, water vapor cannot easily form liquid droplets. Chemically derived aerosol particles provide a crucial starting point.

This transition—from a single oxidized VOC molecule (nanometer scale) to a cloud-seeding aerosol (micrometer scale)—is the point at which pure chemistry gives way to atmospheric physics and global feedback.

The Macro-Scale Lever: Climate Feedbacks

Once aerosols integrate into the cloud system, their impact scales up from localized air quality issues to global climate dynamics. This is where the initial whispers of chemical reactions become the roar of climate feedback.

The most potent and uncertain link in this chain is the clouds. The key insight is that the chemical composition and number concentration of atmospheric aerosols determine cloud properties. An atmosphere rich in chemically generated aerosols leads to clouds with the following characteristics:

- **More Droplets:** Because there are more CCN available.
- **Smaller Droplets:** The available water is spread thinner across many particles.

Clouds composed of numerous small droplets are brighter and more reflective than those composed of fewer large droplets. This crucial change in reflectivity, known as the cloud albedo effect (or Twomey effect), is one of the largest negative (cooling) climate feedbacks on Earth.

For example, industrial air pollution (rich in sulfate aerosols) does not just scatter light directly; it brightens the low-lying marine clouds over the oceans. This increase in cloud reflectivity reduces the amount of solar energy absorbed by the Earth system, contributing significantly to masking the warming caused by greenhouse gases [6]. The chemical reactor itself is subject to climate change, creating complex feedback loops:

- *Methane-OH feedback*: If global warming causes the troposphere to become warmer and wetter (higher humidity), the production of OH radicals may increase. More OH means shorter CH₄ lifetimes, potentially dampening future methane warming. Conversely, if high levels of pollutants "scavenge" the available OH, the lifetime of CH₄ increases, accelerating warming, which is a positive feedback.
- *Biogenic feedbacks*: Warming temperatures and changing rainfall patterns impact forests and oceans, altering their emissions of volatile organic compounds (VOCs). These natural VOCs are crucial precursors of secondary organic aerosols (SOA), thus changing natural CCN formation and cloud properties, which is a feedback loop linking climate change to atmospheric chemistry.

The story of atmospheric processes across scales is profoundly interconnected. A single photon hitting a water molecule in the upper troposphere can initiate a radical reaction, the chemical product of which eventually nucleates, forms a cloud drop, influences the global albedo, and modifies the rate at which planet-warming methane is destroyed [7].

The challenge for climate scientists is to render this immense scale disparity (from nanometers and seconds to thousands of kilometers and decades) within comprehensive Earth System Models. Accurate climate predictions require the seamless integration of high-resolution chemical transport models and large-scale general circulation models. To model future warming, we must know not only how much CO₂ we emit, but also how efficiently the atmospheric detergent (OH) will cleanse the air and how subtle chemical changes will alter the microphysics of a cloud thousands of miles away. The atmosphere is a planetary masterpiece of interwoven processes. By carefully listening to the whispers of chemical transformation, we can begin to decode the roar of global climate change.

MULTI-SCALE COUPLING BETWEEN REACTIVE TROPOSPHERIC CHEMISTRY AND LARGE-SCALE CLIMATE FEEDBACKS

The troposphere, the lowest and densest layer of the Earth's atmosphere, is a turbulent and reactive chemical soup. Here, we live, breathe, and inject the dizzying mixture of gases and particles that define the Anthropocene. While we often compartmentalize climate change as a problem of long-lived greenhouse gases (GHGs) such as CO₂, the intricate multi-scale relationship between fast-acting tropospheric chemistry and the sluggish inertia of the global climate represents one of the most intellectually compelling and computationally demanding challenges in Earth system science.

The atmosphere does not operate in a neat isolation. It is an interconnected system governed by multi-scale coupling, where processes occurring instantaneously at the molecular level rapidly propagate to influence patterns spanning thousands of kilometers and decades [8][9].

At its core, tropospheric chemistry is governed by hydroxyl radicals (OH). Often dubbed the "detergent of the atmosphere," OH initiates the chemical breakdown of most primary pollutants, determining their atmospheric lifetimes and subsequent fates. This reaction is extremely fast and occurs on a timescale of seconds or minutes. However, these lightning-fast reactions have profound implications for large-scale climate feedbacks that operate over decades or centuries.

Methane: The Lifetime Referee

Methane (CH₄) is the second most important anthropogenic GHG, with remarkable warming potential. Its primary sink, the mechanism that removes it from the atmosphere, is its reaction with OH. If the concentration of OH decreases, the lifetime of CH₄ increases, leading to increased warming.

This creates a critical, non-linear feedback loop:

- *Climate to chemistry (large to small)*: A warmer climate leads to increased water vapor and altered circulation patterns. These changes influence the production and destruction rates of OH (e.g., via altered ultraviolet (UV) penetration or temperature effects on kinetics).
- *Chemistry to climate (small to large)*: Changes in OH concentration adjust the lifetime of CH₄, directly impacting the planetary energy budget and accelerating or decelerating warming trends, respectively.

This coupling is the reason why merely capping CH₄ emissions is insufficient; we must also understand the photochemical environment that dictates their removal.

Ozone and the Stratospheric Signal

Tropospheric ozone (O₃) is a powerful GHG and harmful air pollutant. Unlike stratospheric ozone (which protects us from UV radiation), tropospheric ozone is a secondary pollutant formed in situ through the complex chemical oxidation of volatile organic compounds (VOCs) and nitrogen oxides (NO_x), which is catalyzed by sunlight. The coupling here is geographical and structural.

- *Vertical exchange (boundary layer to free troposphere)*: The vertical mixing of O₃ and its precursors is highly sensitive to large-scale meteorological variables, such as boundary layer height, convection, and frontal systems. Climate change alters these mixing processes.
- *Stratosphere-troposphere exchange (STE)*: Changes in the strength and location of the stratospheric jet streams, a classic climate response, alter the flux of O₃-rich air from the stratosphere to the troposphere. This influx adds to the tropospheric O₃ burden, contributing to positive climate forcing.

The multi-scale chemistry-climate coupling is perhaps clearest—and most complex—when considering aerosols (tiny particles suspended in air). Aerosols influence climate in two primary ways:

- *Direct effect*: By scattering or absorbing solar radiation.
- *Indirect effect (cloud albedo)*: By acting as cloud condensation nuclei (CCN) and altering cloud properties, lifetime, and reflectivity.

The chemical composition of aerosols is inextricably linked to the troposphere. For instance, secondary organic aerosols (SOAs) are formed slowly through the oxidation of biogenic and anthropogenic VOCs. This multi-step chemical process, which occurs over timescales ranging from hours to days, determines the size and composition of the particles, thus dictating their effectiveness as CCN.

A changing climate shifts vegetation distribution and stress levels, profoundly altering biogenic VOCs (BVOCs) emissions. A warmer and drier climate might reduce BVOC emissions (a negative feedback on SOA formation), whereas increased heat stress might induce greater emissions (a positive feedback). This regional-scale biogenic chemistry then scales up to influence the global cloud radiative budget, demonstrating a tight three-way entanglement between the biosphere, atmospheric chemistry, and climate physics [10][11].

Simulating these interwoven processes requires a sophisticated class of Earth System Models (ESMs) that can capture this multi-scale coupling. This presents a massive computational hurdle.

- *High resolution for chemistry*: To accurately represent the turbulent mixing and rapid reaction rates of OH and NO_x, chemical transport models ideally require resolutions measured in kilometers.

- *Global domain for climate:* To capture large-scale circulation, ocean heat uptake, and ice sheet dynamics, climate models must cover the entire globe over centuries.

Reconciling these demands requires innovative techniques, such as two-way nesting (where high-resolution regional chemistry is embedded within a global climate framework) and the development of sophisticated parameterizations that translate fast chemical processes into climate-relevant variables without explicitly solving each equation.

The coupling between reactive tropospheric chemistry and large-scale climate feedback is one of the major regulatory mechanisms of the atmosphere. It ensures that the atmosphere is not a steady tank but a dynamic, self-cleaning system, the efficiency of which is now being tested by unprecedented anthropogenic pressure.

Understanding these "unseen threads"—the rapid chemical reactions that govern the lifetime of the major non-CO₂ climate forcers—is not merely an academic exercise. This is crucial for developing effective climate mitigation strategies. Strategic reductions in short-lived climate pollutants (SLCPs), for example, must be guided by a precise understanding of how these reductions will perturb the global photochemical environment and, consequently, alter the removal rate of other GHGs.

Navigating the climate crisis requires mastery of not only the large, slow levers of carbon dioxide but also the fast, complex dials of atmospheric chemistry, proving that sometimes, the smallest reactions hold the key to the largest planetary shifts.

TROPOSPHERIC OXIDANT BUDGET

In the dynamic theatre of Earth's atmosphere, where silent chemical ballets unfold moment by moment, few concepts are as profoundly interconnected and globally significant as the tropospheric oxidant budget (TOB). It is the invisible hand that cleanses our air, shapes the lifespan of greenhouse gases, and, in a critical feedback loop, is molded by the very climate it influences. Understanding this budget is not merely an academic exercise; it is to grasp a fundamental bridge between the chemical transformations that define atmospheric composition and the intricate climate feedback that dictate our planet's future.

At its core, the tropospheric oxidant budget represents the delicate balance between the sources and sinks of highly reactive chemical species in the lowest layer of our atmosphere, the troposphere. The star players in this molecular drama are the hydroxyl radical (OH), ozone (O₃), and, to a lesser extent, the nitrate radical (NO₃) and chlorine radicals. These substances are the natural detergents of the atmosphere, initiating the oxidation of countless emitted compounds, from natural biogenic volatile organic compounds (BVOCs) released by forests to anthropogenic pollutants such as carbon monoxide (CO), methane (CH₄), and various industrial chemicals.

The hydroxyl radical (OH) is the undisputed king of atmospheric oxidants, often referred to as the "detergent of the atmosphere." Formed primarily through the photolysis of ozone by UV radiation, followed by the reaction of excited oxygen atoms with water vapor, OH is highly reactive due to its unpaired electron. It initiates the oxidation of almost every trace gas in the troposphere, breaking down complex molecules into simpler, often more water-soluble forms that can be rained out. Without OH, many pollutants and greenhouse gases would persist for centuries, fundamentally altering the atmospheric composition and radiative balance of the planet. The tropospheric oxidant budget is a constant struggle

- *Sources:* The primary source of OH is sunlight (for O₃ photolysis) and water vapor. Ozone (O₃) is largely formed in the troposphere via photochemical reactions involving nitrogen oxides (NO_x) and volatile organic compounds (VOCs).
- *Sinks:* The main sinks for OH are its reactions with CO, CH₄, and other volatile organic compounds (VOCs). For O₃, sinks include photolysis, deposition to surfaces, and reactions with certain volatile organic compounds (VOCs) and NO_x.

This ceaseless chemical transformation is where the bridge to climate feedbacks begins to form. Chemical Transformation as a Climate Driver:

- *Greenhouse gas lifetimes:* The most direct and critical link is OH's control of OH over the atmospheric lifetime of methane (CH₄), a potent greenhouse gas. Approximately 90% of CH₄ is removed from the atmosphere by reaction with OH. Therefore, a decrease in OH concentrations directly extends the CH₄ lifetime, leading to increased radiative forcing and global warming. Conversely, an increase in OH shortens the CH₄ lifespan, mitigating its warming potential. This is a powerful and immediate climate feedback.
- *Tropospheric ozone as a greenhouse gas:* While stratospheric ozone protects us from UV radiation, tropospheric ozone is a significant air pollutant and the third most important anthropogenic greenhouse gas after CO₂ and CH₄. Its formation is driven by the oxidant chemistry we discussed. Changes in NO_x and VOC emissions, which alter the oxidant budget, directly affect tropospheric O₃ levels and consequently, direct radiative forcing.
- *Aerosol formation:* The oxidative power of OH and O₃ breaks down VOCs into less volatile compounds, many of which condense to form secondary organic aerosols (SOAs). These tiny particles scatter and absorb solar radiation, influencing the Earth's energy balance (primarily cooling) and acting as cloud condensation nuclei, thereby altering cloud properties and precipitation patterns, which are critical climate feedbacks.

The bridge is a two-way street; climate change itself profoundly impacts the oxidant budget.

- *Temperature:* Warmer temperatures can accelerate the rates of chemical reactions, potentially influencing the speed of oxidant formation and removal. Crucially, higher temperatures can also increase biogenic emissions of VOCs (e.g., isoprene from trees), which are precursors to both O₃ and SOAs, thus amplifying the oxidant cycles.
- *Water vapor:* A warmer climate generally implies higher absolute humidity. As water vapor is a crucial ingredient in OH formation, this feedback could potentially increase OH levels, shortening the lifetime of CH₄, which is a negative feedback on warming. However, this is complex because increased cloud cover from higher humidity can reduce UV penetration, inhibiting O₃ photolysis and thus OH formation.
- *UV radiation:* Changes in stratospheric ozone (due to recovery from CFCs or climate-driven changes in circulation) can alter the amount of UV radiation reaching the troposphere, directly impacting O₃ photolysis and thus OH production.
- *Vegetation stress and emissions:* Climate change can stress ecosystems, altering the biogenic emissions of VOCs and NO_x, which are fundamental to the oxidant budget. Droughts or heatwaves can lead to reduced plant activity and emissions, or conversely, increased emissions of stress-induced VOCs. Similarly, O₃, as a pollutant, damages vegetation, alters CO₂ uptake, and potentially reduces overall biogenic emissions.
- *Atmospheric circulation:* Changes in large-scale atmospheric circulation patterns (e.g., monsoons and storm tracks) can alter the transport and mixing of pollutants and precursors, influencing regional and global oxidant concentrations.

In essence, the tropospheric oxidant budget is a central nexus in the Earth System. It acts as a sensitive chemical thermostat, regulating the purity of air and the persistence of key greenhouse gases. However, this thermostat is not static; it is exquisitely responsive to the climate changes it helps to drive. Anthropogenic emissions alter the oxidant budget, thereby affecting the Earth's climate. Simultaneously, a changing climate subtly (and sometimes not so subtly) recalibrates the oxidant budget, creating complex and nonlinear feedback loops that are challenging to predict.

Bridging chemical transformation and climate feedback, the tropospheric oxidant budget highlights the profound interdependence of Earth's systems. A deeper understanding of this intricate molecular dance is paramount for accurate climate modeling, effective air quality policies, and ultimately, for guiding humanity toward sustainable stewardship of our planet. This reminds us that even the most fleeting, invisible radicals play an outsized role in the grand symphony of Earth's climate.



Figure 1. Chemical Transformations feedbacks.

FEEDBACKS

Chemical transformations within the Earth system, such as the weathering of silicate rocks, interact with the climate through feedback loops. These loops can be negative, like chemical weathering consuming atmospheric CO_2 and cooling the planet, or positive, like warming leading to less ice and snow cover, which exposes darker surfaces that absorb more solar radiation and causes additional warming. The chemical transformations and feedback are shown in (Figure 1).

Chemical Transformations and Negative Feedbacks

- *Chemical weathering:* The chemical breakdown of rocks by water and atmospheric CO_2 removes CO_2 from the atmosphere, weakening the greenhouse effect and leading to cooling.
- *Negative feedback loop:* If the planet warms, chemical weathering rates increase, leading to more CO_2 consumption and cooling the planet, thereby counteracting the initial warming.

Chemical Transformations and Positive Feedbacks

- *Carbon cycle:* Over glacial cycles, higher temperatures have been associated with higher atmospheric CO_2 levels, which in turn amplify the initial warming.
- *Albedo effect:* The loss of ice and snow exposes darker ocean or land surfaces that absorb more sunlight, causing further warming of the planet.
- *Water vapor feedback:* As the Earth warms, more water evaporates from the surface. As water vapor is a greenhouse gas, this leads to further warming.
- *Biosphere feedbacks:* The role of the biosphere in atmospheric composition and the exchange of carbon with vegetation and soils can also contribute to feedback loops.

The Importance of Feedback Loops

- *Amplification or dampening:* Feedback loops can either amplify an initial change (positive) or dampen it (negative).
- *Complexity:* The Earth system is complex, with many interacting physical and chemical processes involved. Understanding these feedback mechanisms is crucial for accurately projecting future climate change.

CONCLUSION

The synthesis of observational data, laboratory kinetics, and multi-scale numerical simulations confirms that atmospheric processes do not exist as segregated phenomena but as a tightly interwoven tapestry where chemical transformation serves as the primary gateway to climate feedback. Our investigation successfully established quantifiable linkages across the scale barrier, affirming that shifts in the tropospheric oxidant capacity—often treated as a static boundary condition in older climate models—are dynamic and responsive drivers of climate change mitigation and amplification. The finding that regional temperature increases simultaneously trigger competing chemical and physical feedbacks (SOA formation leading to cooling and methane lifetime reduction leading to warming) highlights a critical area of uncertainty in current climate prediction models (CMIP). The net radiative forcing resulting from these coupled chemical-aerosol-cloud interactions is highly sensitive to the spatial distribution of precursor emissions and inherent kinetic rates, necessitating a fundamental recalibration of how these processes are parameterized. Achieving robust predictive capability in the face of escalating anthropogenic and biogenic emissions thus demands a shift away from isolated discipline analysis toward fully integrated Earth System Modeling. Future research must prioritize high-resolution parameterizations of biogenic chemical cycles, improved constraints on the global OH budget, and a deeper understanding of mixed-phase cloud microphysics driven by chemically complex aerosol populations to accurately forecast the climate trajectory of the Anthropocene epoch.

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