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ATMOSPHERIC PHOTOCHEMISTRY: AN IN-DEPTH REVIEW

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ABSTRACT

Atmospheric photochemistry[1-6] constitutes a foundational component of atmospheric science, governing the chemical transformations that regulate air quality, influence climate processes, and determine the composition of both the troposphere and stratosphere. This review examines the fundamental mechanisms of photochemical reactions, highlighting key reactive species such as radicals (e.g., OH, NO_3), ozone (O_3), and volatile organic compounds (VOCs), as well as their roles in driving complex chemical cycles.

Particular attention is given to the influence of anthropogenic emissions—including nitrogen oxides (NO_x), sulfur compounds, and industrial VOCs—which significantly alter photochemical pathways and exacerbate phenomena such as photochemical smog, ozone depletion, and secondary pollutant formation.

The review further explores recent advancements in atmospheric photochemistry, encompassing the development of computational models, remote sensing technologies, and in situ observational techniques, all of which have enhanced the understanding and quantification of photochemical processes. In light of ongoing environmental challenges, the paper also discusses emerging mitigation strategies aimed at reducing the adverse impacts of human activities on atmospheric chemical balance and climate systems.

1. INTRODUCTION

Atmospheric phot ochemistry encompasses the suite of chemical reactions initiated by the absorption of solar radiation by atmospheric constituents. These photo chemically driven processes play a pivotal role in modulating the concentrations of trace gases, aerosols, and particulate matter within both the troposphere[7-18] and stratosphere[19-20], thereby exerting substantial influence on air quality, climate dynamics, and atmospheric composition.

The study of atmospheric photo chemistry is critical for elucidating mechanisms underlying key environmental phenomena such as stratospheric ozone depletion, photochemical smog formation, and the oxidative degradation of greenhouse gases. These processes have profound implications for public health, ecosystem stability, and global climate regulation.

The significance of photo chemistry in atmospheric science lies in its central role in governing the Earth's radiative energy balance and facilitating the chemical cycling of reactive species. As solar photons are absorbed by molecules such as ozone (O_3), nitrogen dioxide (NO_2), and various volatile organic compounds (VOCs), a cascade of photolytic and radical-driven reactions is initiated. These reactions lead to the formation or destruction of climatically and chemically active trace gases, acting as key intermediates in broader environmental processes, including climate change, tropospheric oxidation capacity, and atmospheric pollutant formation.

2. BASIC PRINCIPLES OF PHOTOCHEMISTRY[21-36]

Photo chemistry is driven by the interaction of electromagnetic radiation, primarily ultraviolet (UV) and visible light, with molecules in the atmosphere. When a molecule absorbs light, it

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gains energy, which can result in a variety of outcomes depending on the wavelength and the nature of the molecule. Photochemical reactions in the atmosphere primarily involve:

- Photoexcitation: The absorption of UV light excites molecules to higher energy states, which can lead to chemical reactions.
- Photo dissociation: Absorption of UV radiation can break chemical bonds, producing free radicals or ions.
- Photoionization: High-energy photons can ionize molecules, producing charged species that may further undergo chemical reactions.
- Radiationless Transitions: After absorbing energy, molecules may return to their ground state without emitting radiation, through processes like internal conversion or vibrational relaxation.

The rate at which photochemical reactions occur depends on several factors, including the intensity of the incident radiation, the photo dissociation cross-sections of the molecules involved, the atmospheric conditions, and the presence of catalysts such as ozone or hydroxyl radicals ($OH^{\cdot}$).

3. PHOTO CHEMICALLY ACTIVE SPECIES IN THE ATMOSPHERE[37-43]

Numerous reactive species participate in atmospheric photo chemistry, each playing critical roles in the stratosphere and troposphere. These species control the formation, transformation, and destruction of trace gases, thereby influencing both climate and air quality.

3.1. Ozone (O_3)[44-46]

Ozone is a central component of atmospheric photo chemistry due to its ability to absorb harmful ultraviolet (UV) radiation, especially in the stratosphere, where it forms the protective ozone layer.

Formation (Stratosphere): The Chapman cycle describes the photochemical production and destruction of ozone:

Photo dissociation of O_2 ($\lambda < 240$ nm):

$O_2 + h\nu \rightarrow 2\ O$ (atomic oxygen) $\quad [\lambda < 240\ nm]$

Ozone formation via three-body reaction:

$O + O_2 + M \rightarrow O_3 + M$

M = a third body (usually N_2 or O_2) that carries away excess energy.

Destruction: Photo dissociation of ozone ($\lambda < 320$ nm):

$O_3 + h\nu \rightarrow O_2 + O$ $\quad$ [UV-B, UV-C]

Recombination: $O + O_3 \rightarrow 2\ O_2$

This dynamic equilibrium maintains stratospheric ozone levels under natural conditions. However, catalytic cycles involving NO_x, ClO_x, and HO_x can accelerate ozone destruction.

Anthropogenic ozone depletion: Human-emitted chlorofluorocarbons (CFCs) release chlorine atoms in the stratosphere:

$CFCl_3 + h\nu \rightarrow CFCl_2 + Cl$

$Cl + O_3 \rightarrow ClO + O_2$

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$ClO + O \rightarrow Cl + O_2$

Net equation

$O_3 + O \rightarrow 2O_2$

3.2. Nitrogen Oxides[47-49] ($NO_x = NO + NO_2$)

Nitrogen oxides are key intermediates in both ozone formation (troposphere) and ozone destruction (stratosphere).

Tropospheric NO_x reactions (ozone formation):

NO_2 photolysis: $NO_2 + h\nu \rightarrow NO + O$ $[\lambda < 420\ nm]$

Ozone formation: $O + O_2 + M \rightarrow O_3 + M$

NO reacts with O_3: $NO + O_3 \rightarrow NO_2 + O_2$

This cycle forms ground-level ozone, a major component of photochemical smog in polluted urban air.

Stratospheric ozone destruction (catalytic cycle):

NO_2 photolysis: $NO_2 + h\nu \rightarrow NO + O$

Catalytic cycle:

$NO + O_3 \rightarrow NO_2 + O_2$

$NO_2 + h\nu \rightarrow NO + O$

Net: $O_3 + O \rightarrow 2\ O_2$

NO_x can be transported to the stratosphere from the oxidation of nitrous oxide (N_2O), emitted from soils and agriculture.

3.3. Hydroxyl radical (OH)[50-52]:

The hydroxyl radical is the most important oxidizing agent in the troposphere, often called the atmospheric detergent.

Formation: Ozone photolysis ($\lambda < 320\ nm$):

$O_3 + h\nu \rightarrow O(^1D) + O_2$

Reaction with water vapour: $O(^1D) + H_2O \rightarrow 2\ OH$

$O(^1D)$ = electronically excited oxygen atom.

Role:OH initiates the oxidation of many trace gases:

Methane oxidation: $OH + CH_4 \rightarrow CH_3 + H_2O$

VOC oxidation: $OH + RH \rightarrow R\bullet + H_2O$

These reactions ultimately lead to CO_2, O_3, organic nitrates, and

secondary organic aerosols (SOA).

3.4. Volatile organic compounds (VOCs)[53-56] :

VOCs are emitted from natural sources (e.g., vegetation: isoprene, terpenes) and anthropogenic sources (e.g., fuel combustion, solvents). They participate in complex photochemical oxidation chains.

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Typical VOC oxidation (e.g., isoprene, C_5H_8):

Initiation (reaction with OH): $C_5H_8 + OH + O_2 \rightarrow RO_2\bullet$

Reaction with NO (in polluted air): $RO_2\bullet + NO \rightarrow RO\bullet + NO_2$

$NO_2 + h\nu \rightarrow NO + O$

$O + O_2 \rightarrow O_3$

Ozone is produced during VOC - NO_x reactions.

Formation of SOA:

$RO\bullet \rightarrow$ low-volatility products $\rightarrow$ condensation $\rightarrow$ SOA

4. PHOTO CHEMISTRY IN THE TROPOSPHERE[56-59]

The troposphere, the lowest layer of earth's atmosphere (extending from the surface up to ~8–15 km), is the site of most weather phenomena and human-induced emissions. Photochemical reactions in this region are critically linked to air pollution, oxidant formation, and climate forcing. Driven by solar radiation, these reactions involve a variety of trace gases and radicals, leading to the formation of ozone (O_3), secondary organic aerosols (SOA), and reactive oxidants that affect both air quality and climate dynamics.

Photochemical processes in the troposphere

1. **Formation of ground-level tropospheric ozone:** Unlike stratospheric ozone, which protects life from UV radiation, tropospheric ozone is a harmful pollutant formed by photochemical smog reactions.

Photolysis of nitrogen dioxide (NO_2):

$NO_2 + h\nu \rightarrow NO + O$ $[\lambda < 420\ nm]$

Ozone formation: $O + O_2 + M \rightarrow O_3 + M$

NO titration reaction: $NO + O_3 \rightarrow NO_2 + O_2$

This cycle alone does not produce net O_3. However, in the presence of volatile organic compounds (VOC) or carbon monoxide (CO), radicals are regenerated and ozone accumulates.:

2. **VOC oxidation and smog formation:** VOC emitted from vehicles, industries, and vegetation react with $OH^{\bullet}$ radicals to form organic peroxy radicals ($RO_2^{\bullet}$), which in turn participate in ozone production.

Typical reactions:

$VOC + OH + O_2 \rightarrow RO_2\bullet$

$RO_2\bullet + NO \rightarrow RO\bullet + NO_2$

$NO_2 + h\nu \rightarrow NO + O$

$O + O_2 \rightarrow O_3$

Net result: Formation of O_3 and photochemical smog

3. Formation of hydroxyl radical ($OH^{\bullet}$): $OH^{\bullet}$ is the primary oxidizing agent in the troposphere, initiating the breakdown of pollutants such as CH_4, CO, and VOCs.

Formation via ozone photolysis:

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$O_3 + h\nu \rightarrow O(^1D) + O_2 \qquad [\lambda < 320\ nm]$

$O(^1D) + H_2O \rightarrow 2\ OH^{\bullet}$

4. Methane (CH_4) oxidation – Greenhouse gas removal: $OH^{\bullet}$ initiates methane oxidation, influencing climate and oxidative capacity.

$CH_4 + OH^{\bullet} \rightarrow {}^{\bullet}CH_3 + H_2O$

${}^{\bullet}CH_3 + O_2 \rightarrow CH_3O_2^{\bullet}$

$CH_3O_2^{\bullet} + NO \rightarrow CH_3O^{\bullet} + NO_2$

$NO_2 + h\nu \rightarrow NO + O$

$O_2 + O \rightarrow O_3$

Leads to ozone formation, contributing to warming despite CH_4 removal.

5. Secondary organic aerosol (SOA) formation[60-79]: Low-volatility products from VOC oxidation condense into particulate matter, forming SOA, which affect radiation balance and cloud properties.

Example pathway (monoterpene):

$C_{10}H_{16} + O_3 \rightarrow$ oxidation products $\rightarrow$ SOA (via nucleation/condensation)

6. NO_x Cycling and nighttime chemistry: At night, NO_3 and N_2O_5 form and participate in heterogeneous reactions.

Reactions: $NO_2 + O_3 \rightarrow NO_3 + O_2$

$NO_3 + NO_2 \rightleftharpoons N_2O_5$

$N_2O_5 + H_2O$ (on aerosol) $\rightarrow 2\ HNO_3$

Leads to nitrate aerosol formation and acidification of precipitation.

Smog formation[80-82] – Atmospheric photo chemistry

Photochemical smog forms in the troposphere when nitrogen oxides (NO_x) and volatile organic compounds (VOC) react under the influence of sunlight (UV radiation). The result is a complex mixture of ozone (O_3), peroxyacyl nitrates (PAN), secondary organic aerosols (SOA), and other oxidants—collectively termed secondary pollutants.

Step wise mechanism of smog formation

1. NO_x Photolysis and ozone formation

   Emissions from vehicles, power plants, and industrial sources release NO and NO_2 into the atmosphere. The main reactions

Photolysis of NO_2:

$NO_2 + h\nu \rightarrow NO + O \qquad [\lambda < 420\ nm]$

Ozone formation via O-atom recombination:

$O + O_2 + M \rightarrow O_3 + M$

Titration of O_3 by NO (reversible step):

$NO + O_3 \rightarrow NO_2 + O_2$

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This cycle alone does not lead to net ozone accumulation unless radicals (e.g., $RO_2^{\bullet}$) from VOC oxidation are involved.

2. VOC Oxidation and radical propagation

VOC from vehicle exhaust, industrial solvents, biomass burning, and natural sources like trees react with $^{\bullet}OH$ radicals, initiating a chain of reactions that generate peroxy radicals ($RO_2^{\bullet}$).

Initiation – $^{\bullet}OH$ attack:

$RH\ (VOC) + OH + O_2 \rightarrow RO_2^{\bullet} + H_2O$

NO reaction with $RO_2^{\bullet}$: $RO_2^{\bullet} + NO \rightarrow RO^{\bullet} + NO_2$

NO_2 photolysis again produces O_3: $NO_2 + h\nu \rightarrow NO + O$

$O + O_2 \rightarrow O_3$

Net result: Multiple molecules of O_3 are formed for every NO_x–VOC cycle.

3. Formation of other smog components

Peroxyacyl nitrate (PAN)[83-98]
Formed from the reaction of acyl peroxy radicals with NO_2:

$CH_3C(O)O_2^{\bullet} + NO_2 \rightleftharpoons CH_3C(O)O_2NO_2$ (PAN)

PAN[99-101] is a major eye and respiratory irritant, and acts as a NO_x reservoir.

Secondary organic aerosols (SOA):
Low-volatility products from VOC oxidation condense to form aerosols, contributing to smog and reducing visibility.

Health and environmental impacts[102-106]

Ozone (O_3): Irritates the lungs, aggravates asthma, and reduces lung function.

PAN and NO_2: Cause eye irritation and respiratory problems.

SOA and $PM_{2.5}$: Penetrate deep into lungs; linked to cardio vascular and respiratory diseases.

Vegetation damage[107-108] and reduced crop yield due to ozone exposure.

4.2. I**mpact of photo chemistry on greenhouse gases[109-128]**

Atmospheric photo chemistry plays a critical role in both the removal and transformation of greenhouse gases (GHG). In particular, the hydroxyl radical (OH) acts as a primary atmospheric oxidant, initiating the degradation of several GHG, such as methane (CH_4) and volatile organic compounds (VOC). Although this process reduces the atmospheric lifetime of these gases, it can also lead to the formation of secondary greenhouse gases such as ozone (O_3) and carbon monoxide (CO), thereby affecting the overall radiative forcing.

1. Methane (CH_4) degradation in the troposphere

Methane is removed primarily via reaction with OH radicals, which are generated through ozone photolysis in the presence of water vapor.

Step wise reaction mechanism:

Step 1: $^{\bullet}OH$ formation (initiated by photolysis of O_3)[129-133]

$O_3 + h\nu \rightarrow O(^1D) + O_2 \qquad [\lambda < 320\ nm]$

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$O(^1D) + H_2O \rightarrow 2\ ^{\bullet}OH$

Step 2: Methane oxidation

Initiation: $CH_4 + {}^{\bullet}OH \rightarrow CH_3^{\bullet} + H_2O$

Peroxy radical formation: $CH_3^{\bullet} + O_2 \rightarrow CH_3O_2^{\bullet}$

Reaction with NO: $CH_3O_2^{\bullet} + NO \rightarrow CH_3O^{\bullet} + NO_2$

Formation of formaldehyde and CO:

$CH_3O^{\bullet} + O_2 \rightarrow HCHO + HO^{\bullet}$

$HCHO + h\nu \rightarrow H^{\bullet} + HCO^{\bullet}$

$HCO^{\bullet} + O_2 \rightarrow CO + HO_2^{\bullet}$

CO Oxidation to CO_2:

$CO + OH \rightarrow CO_2 + H^{\bullet}$

Ozone formation as a byproduct

The NO_2 produced in Step 3 undergoes photolysis:

$NO_2 + h\nu \rightarrow NO + O$

$O + O_2 \rightarrow O_3$

Thus, the oxidation of methane can indirectly increase ozone concentrations in the troposphere, especially in the presence of NO_x.

While CH_4 has a global warming potential ~28–34 times that of CO_2 (over 100 years), its photochemical degradation products also contribute to net warming, though with different lifetimes and impacts.

Photochemical processes in the stratosphere[134-157]: The stratosphere (approximately 10–50 km altitude) hosts the ozone layer, which absorbs harmful ultraviolet (UV-B and UV-C) radiation, shielding life on Earth. The stratospheric chemical system is governed by photolytic reactions, especially those involving ozone (O_3) and trace gases, and maintains a dynamic equilibrium between ozone formation and destruction.

5.1. Ozone formation and natural destruction (Chapman Cycle)[158-168]

formation of ozone (O_3):

Photo dissociation of molecular oxygen: $O_2 + h\nu \rightarrow 2\ O \quad [\lambda < 240\ nm]$

Formation of ozone via three-body reaction: $O + O_2 + M \rightarrow O_3 + M$

M = a third body (usually N_2 or O_2) that absorbs excess energy.

Natural destruction of ozone:

Photolysis of ozone: $O_3 + h\nu \rightarrow O_2 + O \quad [\lambda < 320\ nm]$

Recombination with atomic oxygen: $O + O_3 \rightarrow 2\ O_2$

This set of reactions describes the Chapman mechanism, which maintains the stratospheric ozone balance under natural conditions.

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5.2. Ozone depletion – Anthropogenic impact

Human-made substances such as chlorofluorocarbons (CFC), halons, and carbon tetrachloride (CCl_4) have significantly disrupted this balance.

Mechanism of ozone destruction by CFC and halons[169-181]:

Step 1: Photo dissociation of CFC in stratosphere

CFC are chemically stable in the troposphere but undergo photolysis in the stratosphere:

$CFCl_3 + h\nu \rightarrow CFCl_2 + Cl \qquad [\lambda < 220\ nm]$

Step 2: Catalytic Destruction of O_3 by Cl Radicals

Cl reacts with ozone: $Cl + O_3 \rightarrow ClO + O_2$

ClO reacts with atomic oxygen: $ClO + O \rightarrow Cl + O_2$

Net Reaction: $O_3 + O \rightarrow 2\ O_2$

A single Cl atom can destroy up to 100,000 ozone molecules before being deactivated.

Similar reaction for bromine (from halons):

Halons release bromine atoms (Br) upon photolysis:

Halon-1211 ($CBrClF_2$) + $h\nu \rightarrow$ Br + radicals

Catalytic cycle: $Br + O_3 \rightarrow BrO + O_2$

$BrO + O \rightarrow Br + O_2$

Net: $O_3 + O \rightarrow 2\ O_2$

Bromine is ~60 times more efficient than chlorine at destroying ozone on a per-atom basis.

5.3. Polar ozone holes

Role of Polar stratospheric clouds (PSC)[182-206]:

In the antarctic winter, stratospheric temperatures drop below −78°C, forming PSC, which provide surfaces for heterogeneous reactions that convert inert chlorine reservoirs (e.g., HCl, $ClONO_2$) into photochemically active chlorine (Cl_2).

Heterogeneous reactions on PSC:

$ClONO_2 + HCl$ (on PSC) $\rightarrow Cl_2 + HNO_3$

When sunlight returns in spring, photolysis of molecular chlorine leads to rapid ozone destruction:

$Cl_2 + h\nu \rightarrow 2\ Cl \qquad [\lambda < 400\ nm]$

$Cl + O_3 \rightarrow ClO + O_2$

$ClO + O \rightarrow Cl + O_2$

Net: $O_3 + O \rightarrow 2\ O_2$

As a result rapid depletion of stratospheric ozone over the antarctic, forming the ozone hole. Similar but less intense, processes can occur in the arctic due to typically higher temperatures.

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CONCLUSION

Atmospheric photo chemistry is a critical field of study for understanding the Earth's atmospheric processes and their environmental implications. From the regulation of ozone concentrations to the formation of pollutants like smog and the breakdown of greenhouse gases, photochemical reactions shape the composition of our atmosphere in profound ways. Ongoing research and technological advances continue to reveal new insights into the complex interplay of these processes, informing strategies for mitigating environmental problems such as air pollution and climate change.

As the global community continues to address the challenges of climate change and environmental degradation, a deeper understanding of atmospheric photo chemistry will be essential for developing effective solutions to protect the atmosphere and human health.

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