# Impact of Iodine Oxides on Troposphere

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*Abstract*—**Ozone plays a crucial role in atmosphere by absorbing harmful ultraviolet rays coming from the sun and maintaining the heat budget. Iodine after escaping from the surface of the ocean in to atmosphere has an important impact on atmosphere by reacting with ozone in troposphere. Recent studies shows that photolysis of iodine containing molecular species such as $CH_3I$, $CH_2I_2$, $CH_2ClI$, $I_2$ and HOI may convert into IO by reacting with ozone in troposphere, which has been observed in MBL, polar regions and free troposphere and lower stratosphere. Atmospheric chemistry of iodine is significant for a number of reasons. The first way is that iodine species contribute to ozone depletion through catalytic cycles by changing the concentration of hydroxyl radicals (OH) in the atmosphere, which may affect the oxidizing chemistry of the atmosphere. The change in oxidizing capacity of atmosphere may results into environment pollution and climate change. Second, it is hypothesized that polar iodine species may promote the oxidation of elemental mercury to reactive gaseous mercury compounds.**

**This might be deposited in snowpack, while some part of the gaseous mercury reduced and goes back to the atmosphere, but the remaining part run off in melted water and may enter in to the food chain. Entry of mercury in food chain may cause serious effects on human health and other living organisms. Third, the production of new particles in atmosphere is also an important aspect of the iodine species. These newly formed particles may convert into cloud condensation nuclei (CCN), which can alter the atmosphere's radiative balance and, consequently, the climate. Iodine monoxide produced iodine dioxide (OIO) after self combination in MBL. Higher order iodine oxides ($I_xO_y$) are created by recombining iodine oxide (IO) and iodine dioxide (OIO), and in a dry atmosphere, they finally produce an aerosol with an $I_2O_5$ composition. These iodine oxide particles (IOPs) may act as ice nucleation particles (INPs) and by involving in new particle formation (NPF), which then may affect the cloud formation processes and radiation budget.**

*Index Terms*—**IOPs, ozone, nucleation, cloud condensation nuclei (CCN), climate.**

## I. INTRODUCTION

Oceans are the primary sources of iodine. Volatile organic iodine (VOIs) may arise as a result of photochemical activities at the ocean surface, as iodide ($I^-$) present in surface water came in contact with ozone to produce diiodine ($I_2$) into atmosphere (Garland et al., 1980).

$$H^+ + I^- + O_3 \rightarrow HOI + O_2 \qquad (R_1)$$

$$H^+ + HOI + I^- \rightarrow I_2 + H_2O \qquad (R_2)$$

High energy ultraviolet radiation volatilize more reactive polyhalogenated compounds like diiodomethane ($CH_2I_2$), bromoiodomethane ($CH_2IBr$), and chloroiodomethane ($CH_2ICl$), as well as monohalogenated organic compounds like methyl iodide ($CH_3I$), ethyl iodide ($C_2H_5I$), and propyl iodide (1- and 2-$C_3H_7I$). Diiodomethane ($CH_2I_2$), chloroiodomethane ($CH_2ClI$), and iodoform ($CHI_3$) are the products of the reaction between dissolved organic matter and hypoiodous acid (HOI), formed by the reaction between ozone and iodide ion, which means marine atmosphere is subsequently exposed to these substances (Wang et al., 2014). These iodinated compounds quickly photo dissociate and produce iodine atoms.

These iodine atoms may have a potential to influence the tropospheric chemistry of the atmosphere. Subsurface observations of such species may underestimate their sea to air flow because recent research has shown additional chemical /photochemical processes for the delivery of reactive halogens from the ocean surface to the lower atmosphere. It was observed that when air $O_3$ directly oxidizes $I_2$ in the sea surface microlayer, reactive organoiodine compounds such as $CH_2I_2$, $CHICl_2$, $CH_3I$, and probably $I_2$ are produced (Martino et al., 2009). It is currently unknown if solar irradiance promotes these reactions, which occur in the dark when seawater is exposed to $O_3$, due to the complicating aspect that polyhalogenated organoiodine compounds are very photolabile.

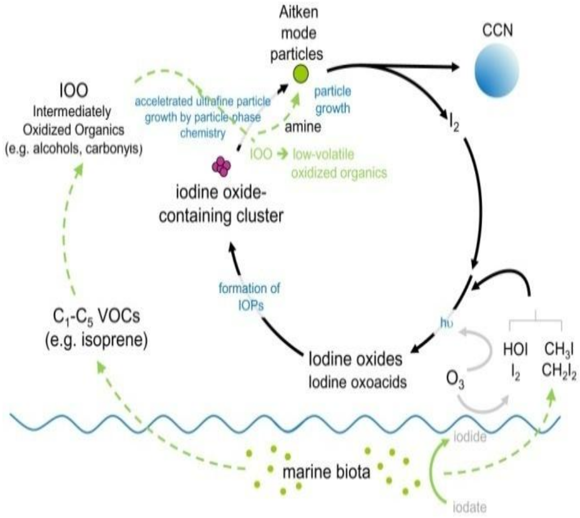

Figure 1: (Source: Huang et al., 2022) Understanding iodine-organic multiphase interactions accelerates the synthesis of new marine particles.

Another potential mechanism to control the synthesis of tiny halogen compounds on the ocean surface is the oxidation of halogen anions to their radical forms by photosensitizers such as chlorophyll or aromatic ketones, a recognized component of marine DOC. When organic compounds are present in the sea surface microlayer, this will lead to the creation of organic halogens (Jammoul et al., 2009; Resser et al., 2005). Ozone behaves as an electron acceptor in atmosphere and encourages the oxidation of atmosphere. Although laboratory data indicates that this should be higher in regions where high concentration of dissolved organic carbon (DOC) exists. The amount of molecular and organic halogen emissions from the open ocean via this route is still unknown.

Iodine in the marine boundary layer is rapidly oxidized after emission (such as $I_2$, HOI, $CH_2I_2$, $CH_3I$, etc.) to form IO and **OIO** radicals. These radicals undergo self reaction pathways such as:

$$\mathrm{IO + IO \rightarrow I_2O_2} \qquad (R_3)$$

$$\mathrm{IO + OIO \rightarrow I_2O_3, I_2O_4}\ \text{etc.} \qquad (R_4)$$

Self-combination of iodine oxides (IO) or the combination of IO and OIO may produce iodine oxides ($I_2O_x$) which then convert in to iodine oxide particles (IOPs). This mechanism has been observed in coastal and polar regions where iodine emissions are strong by absorbing outgoing longwave radiation. The troposphere is being heated by the positive radiative force produced by tropospheric ozone. Ozone is a target for short-term climate mitigation because it has a lifetime of weeks to months and reacts rapidly to changes in precursor emissions. Thus modifying OH levels, ozone formation chemistry controls the radiative forcing of the two gases and influences the atmospheric persistence of methane (Iglesias-Suárez et al., 2018). The impacts of ozone on the environment are greatly influenced by altitude and geographic locations. Because longwave radiation is so sensitive in the high troposphere, ozone has a disproportionately significant warming effect there (Stevenson et al., 2013).

## II. DEPLETION OF OZONE IN TROPOSPHERE

Tropospheric ozone ($O_3$) works as a major greenhouse gas and a dangerous air pollutant in the Earth's atmosphere. Ozone in the lower atmosphere is a secondary pollutant produced by photochemical reactions combining nitrogen oxides ($NO_X$), volatile organic compounds (VOCs), carbon monoxide (CO), and sunlight, in contrast to stratospheric ozone ($O_3$), which shields life by absorbing UV rays.

$$\mathrm{NO + h\nu \xrightarrow{(\lambda=240-320nm)} O\,(3P) + NO} \qquad (R_5)$$

$$\mathrm{O\,(3P) + O_2\,(+M) \rightarrow O_3(+M)} \qquad (R_6)$$

High concentration of ozone in troposphere may be linked with asthmatic problems, allergy, reduced lung functioning and respiratory irritation in human being (Khatri et al., 2009). Ozone easily penetrates plant stomata and induces oxidative stress, which reduces photosynthesis, stunts growth, and reduces crop output (Ainsworth, 2012). It is among the air contaminants that have the greatest negative effects on agricultural productivity worldwide. Ozone influences the oxidation of numerous atmospheric trace gases by acting as a precursor to hydroxyl radicals (OH) through photolysis.

Removal of hydroperoxy ($HO_2$) radicals: HOx refers to the set of highly reactive odd hydrogen radicals, mainly HO and $HO_2$, which control the oxidizing capacity of the troposphere**.** HOx radicals drive oxidation of CO, $CH_4$ and volatile organic compounds (VOCs) and regulate ozone production (McFiggans et al., 2000). Since most daytime OH originates from ozone photolysis (Mahajan et al., 2009):

$O_3 + h\nu \rightarrow O(1D) + O_2$ ($R_7$)

$O\,(1D) + H_2O \rightarrow 2OH$ ($R_8$)

$HO + CO/VOC \rightarrow RO_2 \rightarrow HO_2$ ($R_9$)

$HO_2 + NO \rightarrow HO + NO_2$ ($R_{10}$)

The fundamental basis of atmospheric photochemistry is made up of these cycles. Iodine participates in incredibly quick radical reactions that lower ozone, inhibit $HO_X$, and change radical cycling in the direction of termination. Molecular iodine ($I_2$), hypoiodous acid (HOI), iodocarbons (such as $CH_3I$ and $CH_2I_2$), and other iodine organics are released from coastal and open ocean sources. These species originate from sea-surface chemistry, macroalgae, phytoplankton, and snowpack/surface photochemistry in polar regions (Saiz-Lopez et al., 2012). Iodine atoms produced by the photolysis of above given iodine ($I_2$) or its compounds as given below:

$I_2 \text{ or } RI \rightarrow I + \text{products}$ ($R_{11}$)

(Where R is an organic group)

Iodine atoms produced in this way may react rapidly with ozone present in troposphere.

$I + O_3 \rightarrow IO + O_2$ ($R_{12}$)

This catalytic ozone destruction cycle is well established by (Saiz-Lopez et al., 2008). The most important reaction for the removal of hydrogen peroxide radicals ($HO_2$) from the troposphere is given below:

$HO_2 + IO \rightarrow HOI + O_2$ ($R_{13}$)

This is a strong $HO_2$ sink directly reducing HOx cycling, as demonstrated in laboratory and modeling studies (Saiz-Lopez & von Glasow, 2012), Because $HO_2$ is needed to regenerate OH in $NO_X$ rich air, this step reduces total HOx and slows ozone production cycles (McFiggans et al., 2000)

Removal of hydroxyl (HO) radicals-

OH reacts rapidly with iodine compounds:

$HO + HOI \rightarrow \text{Products}$ ($R_{14}$)

$HO + IO \rightarrow \text{Products}$ ($R_{15}$)

These reactions reduce the hydroxyl (OH) radicals and thereby decrease oxidizing capacity in the marine boundary layer (MBL) (Saiz-Lopez et al., 2014). Field observations at coastal areas confirm that the presence of iodine oxide (IO) correlates with reduced HOx concentrations **(**Stone et al., 2018). Since most daytime hydroxyl (OH) radical originates from ozone photolysis:

$O_3 + h\nu \rightarrow O\,(^1D)$ ($R_{16}$)

$O(^1D) + H_2O \rightarrow 2\,OH$ ($R_{17}$)

A decrease in ozone means less OH formed, leading to an overall decrease in HOx. Thus iodine-driven ozone loss indirectly reduces the main HOx production pathway.

## III. IMPACT ON NOX LEVELS

The combustion of fossil fuels in vehicles, power plants and industrial units is the main anthropogenic, while lightening, soil emissions, biomass burning are the natural sources of nitrogen oxides (Miyazaki et al., 2012). $NO_X$ is a combination of nitric oxide (NO) and nitrogen dioxide ($NO_2$), other oxides of nitrogen like ($NO_3$) are also included in this category (Breuninger et al., 2012). The lifetime of $NO_X$ in the troposphere is relatively short (hours to days), depending on radical concentrations, photolysis rates, and sinks. These species are central to atmospheric radical chemistry and strongly influence ozone ($O_3$), hydroxyl radical (OH), and secondary aerosol production (Miyazaki et al., 2012). During daytime, NO reacts with ozone to form $NO_2$ –

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (R_{18})$$

$NO_2$ may be photolyzed under solar radiation:

$$NO_2 + h\nu \rightarrow NO + O \qquad (R_{19})$$

The atomic oxygen (O) generated can then react with $O_2$ (plus a third body) to regenerate ozone, closing the cycle.

$$O + O_2 + M \rightarrow O_3 + M \qquad (R_{20})$$

This cycle is often called the photo stationary state of $NO_X$ and ozone ($O_3$) (Ravina et al., 2022). NOX interacts with other radicals ($HO_X$, $RO_2$, etc.). For example,

$$HO_2 + NO \rightarrow OH + NO_2 \qquad (R_{21})$$

$NO_2$ can also form nitric acid ($HNO_3$) via reaction with OH. This is a major sink of $NO_X$:

$$NO_2 + OH \rightarrow HNO_3 \qquad (R_{22})$$

This reaction leads to the formation of nitrate aerosol. At night, $NO_X$ can be part of other radical cycles, e.g., $NO_3$ radical chemistry, dimerization, heterogeneous processes, etc. $NO_X$ regulates tropospheric ozone both its formation and destruction. It also influences the oxidizing capacity of the atmosphere because of its interaction with $HO_X$ (HO, $HO_2$) and other radicals. Through nitric acid formation and nitrate aerosols, $NO_X$ affects air quality (acid rain, particulate matter) and climate. Sherwen et al., (2016) investigated the effects of iodine chemistry on tropospheric oxidants in a global model (GEOS-Chem).

By changing NO/$NO_2$ ratios, producing iodine nitrates, and influencing recycling, they discovered that iodine can significantly affect $NO_X$ levels. In particular, regional shifts in $NO_X$ mixing ratios caused by iodine-catalyzed chemistry can have an impact on the ozone and $HO_X$ cycles. When iodine chemistry was included in a modeling experiment over the northern Indian Ocean, $NO_X$ changed significantly. The paper states that when iodine chemistry is active, $NO_X$ concentrations in some areas of the marine boundary layer can be reduced by up to 50% (Mahajan et al., 2021). The process is that the NO/$NO_2$ ratio is shifted when iodine oxides combine with NO to create $NO_2$. Additionally, on aerosol surfaces, iodine nitrates (such $IONO_2$) may arise and be assimilated. This heterogeneous uptake acts as a sink (or sometimes recycling) for both iodine and nitrogen species. Saiz-Lopez et al., (2016) proposed and modeled a new nighttime reaction

$$NO_3 + HOI \rightarrow IO + HNO_3 \qquad (R_{23})$$

## IV. OXIDATION OF MERCURY BY IODINE

The fate of mercury, a pollutant that is present everywhere in the planet, is mostly determined by oxidation-reduction processes. Elemental mercury ($Hg^0$), the most common atmospheric form, has a residence time of numerous months until it is converted into more soluble oxidized forms ($Hg^{II}$), which deposit rapidly in terrestrial and aquatic ecosystems. Halogens are the primary oxidants of atmospheric mercury. Through reactions involving atomic iodine (I) and iodine monoxide (IO), iodine directly influences the oxidation of gaseous elemental mercury. $Hg^0$ may reacts with I to form HgI and with IO to produce HgO and regenerate iodine atoms (I), according to theoretical research and laboratory kinetics. These processes offer a feasible route for the oxidation of mercury in the free troposphere and marine boundary layer due to iodine (Steffen et al., 2008).

Mercury depletion events (MDEs) cause rapid oxidation and deposition of mercury in the Arctic and Antarctic springs. Strong halogen activation from saline snow and sea ice surfaces coincides with these occurrences. Although bromine radicals (BrO) dominate MDE chemistry, observations usually show higher IO concentrations during these periods. By interacting with bromine activation pathways and contributing to increased ozone loss, iodine indirectly raises the rates of mercury oxidation. Iodine can account for a quantifiable fraction of Hg oxidation during MDEs, and iodine–bromine interactions have a major impact on the timing and intensity of these events, according to modeling studies by (Zhang et al., 2012).

Global photochemical modeling has recently shown that iodine radicals are present at quantities high enough to affect mercury oxidation in both the free troposphere and the marine boundary layer. Iodine's impact is not insignificant, even though it is still less than bromine's, especially in tropical areas with high oceanic iodine emissions. The significance of iodine mercury interactions extends beyond coastal and polar habitats, as demonstrated by Saiz-Lopez et al., (2008), who demonstrate that iodine chemistry can influence mercury oxidation pathways at elevations where bromine concentrations are lower.

When combined, these findings show that iodine functions as an indirect regulator of the halogen radical environment that controls mercury's atmospheric lifetime as well as a direct oxidant of elemental mercury. Despite being less common than bromine, iodine plays a significant role in the cycling of mercury, particularly in marine and polar atmospheres, due to its quick photochemistry, high ozone depleting efficiency, and contribution to particle generation. Iodine can change the speciation and destiny of mercury through interactions on iodine rich aerosols, increase the oxidation of mercury, and improve deposition during polar mercury depletion episodes. Iodine's contribution to atmospheric mercury cycling is anticipated to grow in importance as oceanic iodine emissions rise in response to global shifts in ozone and temperature.

Formation of iodine oxide Particles (IOPs): Iodine (as $I_2$, HOI, $CH_2I_2$, $CH_3I$, etc.) in the marine boundary layer (MBL) is rapidly oxidized to IO and OIO radicals through photolysis.

$$I_2, HOI, CH_3I, CH_2I_2 \rightarrow \text{Photolysis} \rightarrow I \qquad (R_{24})$$

$$I + O_3 \rightarrow IO + O_2 \qquad (R_{12})$$

IO reacts with itself (IO + IO) to give OIO, $I_2O_2$, $I_2O_4$, etc. (O'Dowd, et al., 2002)

$$IO + IO \rightarrow I_2O_2 \qquad (R_{25})$$

$$IO + OIO \rightarrow I_2O_3 + I_2O_4 \text{ etc} \qquad (R_{26})$$

These iodine oxides may further reacts with NO, $HO_2$ and other radicals. Formation and growth of iodine oxides (OIO, $I_2O_X$) may lead to the new particle formation (NPF) and aerosol production in marine/coastal atmospheres. IO is therefore both short-lived radical destroying ozone and a precursor to iodine oxide particles (Allan et al., 2015; He et al., 2023).

$$IO + IO / IO + RO_2 / IO + NO \rightarrow OIO / I_2 / I_2O_X \rightarrow \text{particle formation (NPF)} \qquad (R_{28})$$

Growth to Cloud Condensation Nuclei (CCN): When nucleation occurs, iodine oxide particles (IOPs) are initially relatively small (1-2 nm). According to Allan, J. D., et al. (2015), iodine-driven new particle formation events in the Arctic contribute to aerosols that, in specific circumstances, can become CCN. They must develop to around 50–100 nm in order to function as cloud condensation nuclei (CCN) and affect climate. Iodine oxides or iodine oxoacids ($HIO_3$ and $HIO_2$), which can nucleate to generate IOPs or help in the early cluster development by chemically pushing the prenucleation cluster, are produced when iodine released from the ocean surface is photolyzed and oxidized.

Iodine oxide particles (IOPs) formed by the n nucleation are initially very small (1–2 nm). Allan, J. D., et al. (2015) found that iodine driven new particle formation events in the Arctic contribute to aerosols capable of becoming CCN under certain conditions. To influence climate, they must grow to approximately 50–100 nm to act as cloud condensation nuclei (CCN). Iodine released from the ocean surface is photolyzed and oxidized to produce iodine oxides or iodine oxoacids ($HIO_3$ and $HIO_2$), which can nucleate to form IOPs or aid in the early cluster formation by chemically driving the prenucleation cluster. Heterogeneous interactions between the higher iodine oxides and condensing alcohols or carbonyls from the oxidation of marine volatile organic compounds (VOCs) within the newly formed ultrafine particles result in low volatility oxidized organics. These organic acids can combine with basic molecules (such amines) to produce highly hygroscopic salts, which can boost the early particle growth into Aitken mode and ultimately CCN. In order to restart the chemical chain, the higher iodine oxides are recycled during this phase.

Uncertainties in the Climate Impact of IOP Derived Aerosols: Particle formation, halogen radical budgets, and tropospheric ozone loss are all significantly impacted by iodine oxides, mainly IO, OIO, $I_2O_2$, $I_2O_3$, and higher oligomers. There are still a lot of unknowns about how iodine oxides develop, react, and affect climate relevant processes, despite significant advancements over the past ten years. Both global scale atmospheric models and regional chemistry (marine boundary layer, polar areas) are impacted by these uncertainties. Below is a summary of the main knowledge gaps. Despite strong evidence that iodine drives new particle formation, the climate impact remains uncertain for several reasons:

A. Variability in Iodine Emissions: Iodine emissions depend on ozone concentration, sea-surface chemistry, sunlight, biological activity, and halogen activation in sea ice regions. Some studies Shows how sensitive iodine emissions are to climate and ozone changes (Carpenter, et al., 2012).

B. Efficiency of IOP growth to CCN: Not all new particles grow into CCN; many are lost by coagulation. Growth highly depends on availability of condensable vapors ($HIO_3$, organics, $H_2SO_4$ (He et al., 2023).

C. Interactions with other Aerosol Systems: Iodine oxide particles (IOPs) may interact with sulfate, organic aerosols, sea-salt aerosol surfaces. These interactions can suppress or enhance iodine contributions (Saiz-Lopez and von Glasow, 2012).

D. Uncertain Gas Phase Reaction Kinetics and Pathways: Some uncertainties in the gas phase reactions include rate constants for OIO + OIO and IO + OIO reactions, branching ratios between formation of stabilized clusters vs. fragmentation, temperature and humidity dependence of iodine oxide growth, and photolysis rates for OIO and $I_2O_3$ due to limited laboratory spectra. When models try to replicate iodine-driven particle production, this causes a great deal of unpredictability.

E. Iodine Oxide Particle (IOP) Formation and Growth: The mechanics underlying nucleation are still not well understood. Although the production of new particles has been confirmed to occur at high nucleation rates, the relative roles of $I_2O_4$ and $I_2O_5$ in nucleation remain unclear and require further investigation. CCN-sized particles cannot be explained by IOP nucleation alone unless they undergo substantial growth. One of the uncertainties is whether organic condensation will continue to develop IOPs. Whether iodine compounds in the particle phase break back into the gas phase or stay stable. As a result, climate modeling of halogen-induced nucleation is highly unreliable.

F. Heterogeneous Chemistry on Iodine Oxide Surfaces: Other challenges encountered by researchers include the conversion of particulate iodine to volatile organoiodine species and the uptake of HOI, $IONO_2$, $HNO_3$, or ozone regeneration of reactive iodide from aerosols.

G. Uncertainties in Climate Feedback: Although estimates of iodine exposure vary greatly (1–5 ppt), iodine destroys ozone significantly more effectively than chlorine or bromine. There should be significant variations in ozone loss for every small shift in iodine availability. Cloud droplet counts, cloud brightness, and local radiative forcing can all be impacted by IOP formation. However, due to the uncertainty surrounding CCN activation of iodine-rich particles, the effect's magnitude and sign are still unclear.

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