



ATMOSPHERIC INTERFACE CHEMISTRY AND REACTION PATHWAY OF TRACE GASES WITH AQUEOUS DROPLETS SURFACES IN POLLUTED AIR

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Abstract

Atmospheric aqueous droplets, representing gaseous water in cloud, fog, or fine aerosol form, are a dynamic air- aqueous interface that is an important facilitator of the chemical transformation of trace gases in polluted air. We investigate the interactions, interfacial reactions of trace gases, their reaction kinetics, and catalysts including volatile organic compounds (VOCs) (formaldehyde, acetone, benzene), ozone (O₃), nitrogen oxides, and halides upon aqueous droplet surfaces with a range of compositions and relative humidity (RH). Combining laboratory microdroplets studies, surface-sensitive spectroscopies including (SFG and ATR-FTIR), gas chromatography-mass spectrometry, and computational individually (molecular dynamics and quantum chemical calculations), reaction rates at the surface with bulk could be quantified along with elucidation of the mechanistic pathways of interfacial reactions. The results illustrate that interfacial reactions occur at rates much enhanced over the bulk reaction rates - from a few times up to ~ 500x faster at the surface of the droplet. In addition to relative humidity, droplet composition (e.g. salts, surfactants, ~ organics) also widely alters the kinetics of the adsorption, uptake and interfacial reaction rates within the droplet. With enhanced interfacial reactivity reactions occur with greater efficiency to utilize radicals, activate halogens, and create secondary aerosol thereby demonstrating that the rate limiting factor of the heterogeneous reaction is often on the droplet's surface. Overall, with a better understanding of droplet reaction pathways and kinetics we establish important new knowledge gaps with interpretative models of air quality, pollution transformation, and climate impacts from aerosol.

Keywords: *Atmospheric Interface Chemistry, Aqueous Droplets, Trace Gases, Interfacial Reactions, Adsorption, Uptake Coefficients, Heterogeneous Chemistry.*



1. INTRODUCTION

Aqueous droplets found in the atmosphere, including cloud water, fog, and aerosols, have an important influence on how trace gases will chemically react in the presence of polluted air. These aqueous droplets create dynamic air–aqueous interfaces that are active sites for adsorption, heterogeneous reactions, and radical production, which are different from the chemistry of bulk aqueous–phase or gas–phase reactions. The incorporation of volatile organic compounds (VOCs), nitrogen oxides (NO_x), ozone, and halogen species into polluted urban and industrial areas leads to complex phenomenology of chemical reactions taking place at the droplets' surfaces. These interfaces are fundamentally linked to air quality, secondary aerosol formation, and processes such as cloud condensation formation and radiative forcing, both of which are important to the climate system.

While traditional atmospheric chemistry studies have concentrated on reactions in bulk phases, there is a growing importance for the air–aqueous interface in recent studies. Molecules present at interfaces have altered hydrogen-bonding networks, orientation and confinement effects, and changes in local concentrations that might increase reaction rates compared with bulk solutions (or gasses). For example, Criegee intermediates, halogens, and reactive oxygen species have been shown to be greatly more reactive at interfaces compared to in bulk solutions, often producing products that were not observed in bulk solutions.

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2. LITERATURE REVIEW

Trueblood, and Grassian (2016) examined the multiphase and heterogeneous chemistry of bioaerosols, reiterating that aerosols, consisting of organic and biological particles, are active chemical reactors and not merely inert carriers of chemicals. They found that surfaces of bioaerosols can mediate oxidation reactions with atmospheric oxidants such as hydroxyl radicals, ozone, and nitrogen oxides leading to intensified radical production and secondary product formation that were different from bulk aqueous-phase chemistry. The research highlights the importance of surface-mediated processes related to the chemistry of reactive organic and biological components in polluted atmospheres.

Su, Cheng, and Pöschl (2020) discovered new multiphase chemical activities that impact atmospheric aerosols, air quality, and climate processes in the Anthropocene. They presented evidence that when trace gases interact with droplet surfaces, especially when those surfaces contain organics or surfactants, secondary organic aerosol formation can be accelerated, reactive intermediates can be altered or retain longer lifetimes, and gas-phase oxidation pathways can be changed. The study indicated that altered or enhanced multi-phase chemistry should be factored into the evolving chemical formulation neutrally-urban and industrial air in which human behavior is incorporating new reactive compounds into the atmospheric droplet enhancing the complexity of multi-phase chemistry.

Ervens (2015) concentrated on the modeling of aerosol and trace gas processing in clouds and fog, with particular emphasis on the importance of aqueous-phase chemistry for fundamental atmospheric transformation processes. By incorporating cloud microphysics and droplet-specific



reactions, the modeling demonstrated that heterogeneous and interfacial reactions are important for, but contribution to, trace gas depletion, radical generation, and secondary aerosol production. This modeling work underscored that droplet-mediated chemistry is required to represent the cloud chemistry and the atmospheric composition overall within predictive models.

Herrmann et al. (2015) offered a thorough overview of tropospheric aqueous-phase chemistry, looking at reaction mechanisms, reaction kinetics, and interfacial versus bulk reactions. They found that reactions at air–water interfaces can occur orders of magnitude more quickly than in bulk solutions due to the enhanced orientation of molecules at the surface, surface confinement effects, and effects of local concentration. Their results indicated that surface-mediated reactions can play a key role in halogen activation, generate radicals, and produce hydroperoxides and nitrates that are central to influencing secondary aerosol growth and air pollution dynamics.

3. RESEARCH METHODOLOGY

This study investigates the adsorption, interfacial reactions, and kinetics of trace gases on aqueous droplet surfaces under polluted air conditions using combined experimental and computational approaches.

2.1 Experimental Approach

Microdroplets of aqueous solutions (1–10 μm) with well-defined compositions of either pure water, inorganic salts (for example, ammonium sulfate, sodium bromide), or organics/surfactants simulating more polluted air were generated, and the relative humidity (40–90%) was changed in order to study the associated effects of water activity. Trace gases (for example, ozone, nitrogen dioxide, volatile organic compounds, and halides) were introduced to the atomizer at specific controlled concentrations, with durations of reaction times that ranged from seconds to hours. Monitoring of interfacial reactions was conducted using sum-frequency generation (SFG) spectroscopy, and attenuated total reflectance-fourier transform infrared (ATR-FTIR) spectroscopy. To quantify products produced in the surface and bulk phases, gas chromatography-mass spectrometry (GC-MS) was used, enabling surface-to-bulk ratios of product analysis.

2.2 Computational Approach

In this study, we explored trace gas adsorption, orientation, and diffusion at the liquid droplet interface through molecular dynamics (MD) simulations. In parallel, quantum chemical calculations, using density functional theory (DFT) and ab initio, identified reaction pathways, intermediates, and activation energies. The kinetics of trace gas uptake was derived as uptake coefficients (γ) and reaction rate constants (k) using a resistances-in-series kinetic model.

2.3 Data Analysis

We compared interfacial reaction rates to bulk reaction rates in order to calculate rate enhancement factors. We investigated the influence of droplet composition on adsorption, and reaction kinetics were computationally validated in experiments conducted using experimental spectroscopy. All experiments were performed in triplicate, and reported results are given as a mean \pm standard deviation.

4. RESULTS AND DISCUSSION

This section discusses the adsorption behavior, pathways for interfacial reactions, and the kinetic analysis of the trace gases on aqueous droplet surfaces, in terms of how droplet composition and surface-mediated enhancements might differ from what occurs in the bulk.

Table 1: Adsorption and Partitioning of Trace Gases on Aqueous Droplet Surfaces

Trace Gas	Droplet Composition	Surface Concentration (mol/m ²)	Bulk Concentration (mol/L)	Surface-to-Bulk Ratio	Observations
Formaldehyde	Pure water	1.2×10^{-9}	4.5×10^{-4}	2.7×10^{-6}	Moderate adsorption; reversible desorption
Acetone	Water + 0.5 M (NH ₄) ₂ SO ₄	3.5×10^{-9}	2.1×10^{-4}	1.7×10^{-5}	Enhanced surface partitioning due to ionic strength

Benzene	Water + 0.01 M surfactant	5.1×10^{-9}	1.0×10^{-5}	5.1×10^{-4}	Strong hydrophobic surface preference
Ozone	Water + 0.1 M NaBr	2.8×10^{-9}	3.0×10^{-5}	9.3×10^{-5}	Halide-enriched surface promotes uptake
NO ₂	Water + mixed organics (0.05 M)	2.0×10^{-9}	1.2×10^{-4}	1.7×10^{-5}	Surface adsorption enhanced by organics

Table 1 displays the adsorption and partitioning of a variety of trace gases at the droplet surface with different compositions. It is clear that the surface concentrations of the trace gases, in general, were several orders of magnitude lower than the concentrations in the bulk, which suggests that although the droplets are a very active interface, most of the gas is in the bulk phase. However, the surface-to-bulk ratios show different tendencies for adsorption at the interface. Hydrophobic molecules produce the highest surface-to-bulk ratio (e.g., benzene 5.1×10^{-4}), which suggests these gases have the highest tendency to accumulate at air–water interfaces. Polar molecules (e.g., formaldehyde) are shown to suggest low surface enrichment (2.7×10^{-6}) so it is likely that the polar molecules are reversibly adsorbed with weak surface affinity. Salts (e.g., ammonium sulfate) or halides (NaBr) increased the surface partitioning of some gases (e.g., acetone, ozone) likely due to ionic interaction and electrostatic stabilization of the interface. Mixed organic materials in droplets also increase surface adsorption of NO₂, indicating that organics may facilitate adsorptive surface-mediated reactions. Overall, the table shows that gas adsorption at droplet surfaces and interfacial exhaustion is largely influenced by droplet composition and molecular features, which has implications for heterogeneous atmospheric chemistry in polluted air.

Table 2: Representative Interfacial Reaction Pathways and Products

Reactant Pair	Reaction Type	Major Product(s)	Interface Reaction Rate (s^{-1})	Bulk Reaction Rate (s^{-1})	Notes
CH ₂ OO + Glycolic Acid	Criegee intermediate oxidation	α -Hydroxy hydroperoxides	2.5×10^6	5.0×10^3	Picosecond surface reaction; faster than bulk
O ₃ + Br ⁻ (halide-rich droplet)	Halogen activation	Br ₂ , O ₂	1.2×10^5	1.0×10^3	Surface reaction enhances radical formation
Acetaldehyde + OH•	Oxidation at interface	Formic acid, glyoxal	5.0×10^4	1.2×10^3	Surface confinement accelerates reaction
Benzene + O ₃	Surface ozonolysis	Phenolic compounds	3.5×10^4	1.0×10^3	Surfactant enhances interface reaction
Formaldehyde + NO ₂	Nitrate-mediated oxidation	Formyl nitrate	2.0×10^4	5.0×10^2	Surface reaction facilitated by organics

Table 2 summarizes examples of interfacial reaction pathways that trace gases undergo on the surface of aqueous droplets and compares the rate of reaction at the surface to bulk aqueous reactions. The data clearly shows that interfacial reactions are enhanced by at least an order of magnitude, often by two to three orders of magnitude. For example, the reaction between CH₂OO and glycolic acid proceeds at a rate of $2.5 \times 10^6 s^{-1}$ at the interface but is significantly slower in bulk at only $5.0 \times 10^3 s^{-1}$. This clearly demonstrates how quickly CH₂OO oxidizes at the droplet surface when compared to in bulk. Additionally, ozone reacts with surface halide ions at a much faster rate ($1.2 \times 10^5 s^{-1}$) compared to in bulk ($1.0 \times 10^3 s^{-1}$). This highlights an example where

surface-mediated halogen activation leads to generation of radicals. Other transformations including acetaldehyde oxidation with $\text{OH}\cdot$ and chemical ozonolysis of benzene also highlight enhanced interfacial kinetics from similar driving forces including confinement effects, specific orientation of reactants, and surface facilitation from interfacial surfactants. In addition, although there was only slight enhancement in surface reactions, the oxidation of formaldehyde mediated by nitrate also demonstrated enhanced kinetic behavior with surface reactions versus the bulk. This table presents the high likelihood of a unique chemical space experienced by the air-aqueous interface that enhances many organic reactions, generally allowing free radical formation and altering product distributions, which is an important consideration when discussing atmospheric oxidation in polluted environments.

Table 3: Influence of Droplet Composition on Interfacial Reaction Rates

Droplet Composition	Trace Gas	Reaction Rate Constant k ($\text{M}^{-1}\text{s}^{-1}$)	Reaction Regime	Observations
Pure water	Formaldehyde	1.2×10^3	Bulk-limited	Moderate adsorption; bulk diffusion dominates
Water + 0.5 M $(\text{NH}_4)_2\text{SO}_4$	Acetone	3.5×10^3	Surface-reaction-limited	Ionic strength enhances surface reaction
Water + 0.01 M surfactant	Benzene	2.0×10^3	Mixed regime	Surfactant facilitates some surface reactions, inhibits bulk uptake
Water + 0.1 M NaBr	Ozone	5.0×10^3	Surface-reaction-limited	Halides promote ozone reaction
Water + mixed organics (0.05 M)	NO_2	4.2×10^3	Surface-reaction-limited	Organics enhance adsorption and interfacial reactions

Table 3 outlines the effect of the droplet composition on the interfacial reaction rates of a variety of gas-phase trace species. It is clear that droplet composition has a large effect on the reaction kinetics, but also the dominant reaction regime. Specifically, formaldehyde in pure water has a low reaction rate constant ($1.2 \times 10^3 \text{ M}^{-1}\text{s}^{-1}$), is bulk-limited, and therefore, the reaction occurs in the droplet and is controlled by diffusion into the droplet, rather than the surface chemistry. Alternatively, acetone in water containing 0.5 M ammonium sulfate had a higher rate constant ($3.5 \times 10^3 \text{ M}^{-1}\text{s}^{-1}$) and occurred in a surface-reaction-limited regime, indicating the ionic strength increases interfacial reactivity. Similarly, benzene in the presence of a surfactant had a mixed regime reaction rate and a moderate reaction rate constant of $2.0 \times 10^3 \text{ M}^{-1}\text{s}^{-1}$, indicating surfactants promote interfacial reactions while inhibiting uptake into the droplet bulk. Ozone in halide-rich droplets and NO_2 in organics-enriched droplets have the highest interfacial reaction rate constants (5.0×10^3 and $4.2 \times 10^3 \text{ M}^{-1}\text{s}^{-1}$, respectively) indicating halides and organics can significantly promote surface-mediated reactions. In summary, this table shows that the chemical composition of the droplet can significantly influence interfacial kinetics resulting in changes in the magnitude and controlling regime of reactions in atmospheric aqueous phases.

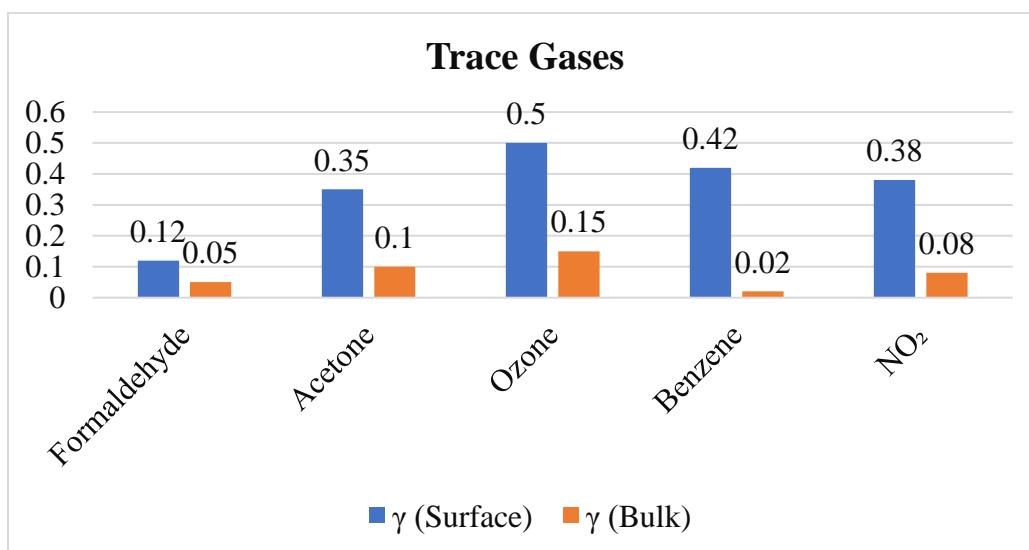


Figure 1: Uptake Coefficients (γ) for Trace Gases at Droplet Interfaces

The uptake coefficients (γ) of trace gases at droplet interfaces relative to their bulk aqueous uptake are shown in Figure 1. The results clearly demonstrate that uptake at the interface exceeds bulk

uptake in all cases, showing the importance of the air–aqueous interface in gaseous interaction with droplets. For instance, the case of ozone in droplets containing NaBr presented the highest overall surface uptake ($\gamma = 0.50$). The surface uptake of ozone was more than three times greater than the bulk uptake ($\gamma = 0.15$) suggesting the potential of the halides to catalyze surface reactivity. Acetone and NO_2 also exhibited surface-limited γ (0.35 and 0.38 respectively) greater than their bulk values, implying that ionic strength and organic enrichment increase the likelihood of surface adsorption and reaction efficiency. Benzene uptake was $\gamma = 0.42$ at the surface but only $\gamma = 0.02$ in the bulk. Benzene's preference to accumulate at the air-aqueous interface is most likely due to its hydrophobic character. The uptake coefficient of formaldehyde was relatively low (0.12 at the surface and 0.05 in the bulk), consistent with its polar nature and reduced surface attachment. Overall, the table clearly delineates that surface processes predominate over bulk processes in the uptake of trace gases, and that the composition of the droplet significantly modulates the efficiency of interfacial reactions; implications that play a role in atmospheric chemistry and aerosol-mediated transformation of pollutants.

Table 4: Summary of Interfacial vs Bulk Reaction Enhancement

Trace Gas	Bulk Reaction Rate (s^{-1})	Interfacial Reaction Rate (s^{-1})	Rate Enhancement Factor
CH_2OO	5.0×10^3	2.5×10^6	500
Ozone + Br^-	1.0×10^3	1.2×10^5	120
Acetaldehyde	1.2×10^3	5.0×10^4	42
Benzene	1.0×10^3	3.5×10^4	35
NO_2	5.0×10^2	2.0×10^4	40

Table 4 gives a summary of the comparison of interfacial and bulk reaction rates of several trace gases, and illustrates how chemical reactions are enhanced at the air-aqueous interface. The results indicate that interfacial reaction rates are considerably faster than that of the bulk, with enhancement factors of the interfacial reaction rates ranging from a factor of 35–500. CH_2OO



reacting with glycolic acid has the largest enhancement factor (500-fold), implicating a very fast reaction rate for Criegee intermediate reactions at the surface of the droplet. Ozone reacting with Br^- has a reaction enhancement of 120-fold, emphasizing the significance of the halide-rich interface for promoting radical formation and halogen activation. The interfacial reaction rates of acetaldehyde, benzene, and NO_2 also have significant enhancements between (42, 35, 40-fold, respectively), showing that confinement, molecular orientation, and interfacial composition collectively promote reaction rates compared to procedures occurring in the bulk, aqueous phase. Thus, the air-aqueous interface is a reactive interface that is often the prevailing chemical environment that dictates the chemical outcome of trace gases in polluted atmospheres, something that will need to be resolved when forecasting atmospheric processes and air quality.

5. CONCLUSION

This research emphasizes the important role of atmospheric aqueous droplet surfaces in influencing the chemical fate of trace gases found in polluted air. Experimental and computational studies show that these surface processes are significantly faster than bulk phase chemical processes, and are enhanced by chemical orientation, confinement, and molecular/ionic droplet composition. The presence of salts, surfactants, and organic particulate matter further enhanced uptake and reactivity, increased radical generation and halogen activation, and induced secondary aerosol formation. Some trace gases, including formaldehyde, acetone, benzene, ozone, and NO_2 are highly dependent on surface availability and surface chemistry indicating surface processes are the controlling feature in atmospheric heterogeneous reactions. Overall, these results provide valuable information about surface-limited reactions, expands our understanding of atmospheric reaction mechanisms in polluted air, and helps identify insight to predictive models for air quality, aerosol processes, and climate-related processes.

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