



Environmental Health Committee (EHC) Emerging Issue Brief

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Indoor Reactive Oxygen and Nitrogen Species

What is the issue?

Reactive oxygen species (ROS) and reactive nitrogen species (RNS) are unstable oxygen- and/or nitrogen-containing radicals and non-radical species. Exposure to extracellular (produced in the environment) ROS and RNS, as well as endogenous (produced inside the body) reactive species production, can result in oxidative stress in humans, which can exacerbate or lead to multiple adverse health impacts, including asthma, diabetes, chronic obstructive pulmonary disease (COPD), and cancer.¹⁻³ ROS and RNS species include superoxide ($O_2^{\cdot-}$), hydroxyl (HO^{\cdot}), hydroperoxyl (HOO^{\cdot}), alkylperoxides (ROO^{\cdot}), hydrogen peroxide (H_2O_2), organic peroxides (ROOR), hypochlorite (OCl^-), peroxyxynitrite ($ONOO^{\cdot}$),⁴⁻⁶ nitrogen oxides (NO_2 , NO ; collectively NO_x), nitrous acid (HONO)⁷, and chlorine nitrite ($ClNO_2$).⁸ Here we focus on indoor extracellular reactive species present in the particle- and gas-phase, rather than intracellular reactive species (generated endogenously).

Indoor Gaseous Reactive Oxygen and Nitrogen Species

Due in part to being a US EPA criteria pollutant (primary annual mean standard of 53 ppb), indoor NO_x is better understood than other ROS species. Ambient NO_x infiltrates into buildings, with penetration factors of about 1 for NO and 0.72 for NO_2 .⁹ In a literature review, median NO_2 concentrations in schools and offices were $26.1 \mu g/m^3$ and $22.7 \mu g/m^3$ with indoor/outdoor ratios of 0.7 and 0.8, respectively.¹⁰ Ventilating combustion appliances and using an outdoor-exhausting stove hood when cooking with natural gas are methods of reducing indoor NO_x emissions.

Gas-phase OH has been measured in a classroom (reaching up to 1.8×10^6 molecules/cm³),^{11,12} during cleaning with limonene (4×10^6 molecules/cm³),¹³ with an electronic air cleaning running (1.8×10^7 molecules/cm³),¹³ and cooking ($2-6 \times 10^6$ molecules/cm³).⁷ In comparison, outdoor OH concentrations range from $2-10 \times 10^6$ molecules/cm³ at midday.⁷ The House Observations of Microbial and Environmental Chemistry (HOMEChem) campaign conducted extensive measurements of the relationships between OH, NO_x , HONO, and O_3 , highlighting the importance of OH production by photolysis of HONO.⁷ NO_2 is a precursor species to indoor HONO formation.^{11,14-16} Combustion is a significant indoor source of NO_x and HONO, including unvented combustion appliances (e.g., gas stoves), smoking, and kerosene heaters.¹⁷ For example, NO_x and HONO concentrations increased from 15 and 4 ppb to 135 and 40 ppb, respectively, during cooking with oil on a natural gas stove.¹⁴ When cleaning with chlorine-based

solutions, reactive chlorine species can be produced through reactions with OH.⁸ Photocatalytic paints can reduce NO₂ concentrations, though significant HONO concentrations may be generated as a result.¹⁸

Indoor Particle-Bound Reactive Oxygen Species

Measurement of particle-bound ROS often employs fluorescent probes calibrated with H₂O₂, with results reported as equivalent nmol/m³ of H₂O₂.¹⁹ While studies of indoor ROS are few, Khurshid et al. (2014) measured concentrations of ROS on PM_{2.5} averaging (±standard deviation) 1.37±1.2 nmol/m³ across twelve residences, with a range of 0.18-4.01 nmol/m³. Similar averages and ranges of ROS on PM_{2.5} have been measured in six institutional buildings (1.16±0.38 nmol/m³, range of 0.63-1.68 nmol/m³), five retail buildings (1.09±0.93 nmol/m³, range of 0.02-3.36 nmol/m³), a university building (3 nmol/m³)²⁰, and six residences (0.90±0.16 nmol/m³, range of 0.40-1.50 nmol/m³)⁶. Compared to traditional approaches, advances in real-time ROS detection have improved measurement accuracy and enabled understanding the dynamics of ROS transport, production, and removal indoors.⁴ Using real-time instrumentation, ROS on PM_{2.5} averaged 2.44±0.40 nmol/m³ and gas-phase ROS averaged 1.80±0.99 nmol/m³ in an unoccupied St. Louis, MO, USA test home.⁴ Additional research is required to understand the relationship between ROS on PM_{2.5} concentrations and health outcomes.

Simultaneous measurements of indoor and outdoor ROS on PM_{2.5} by Khurshid et al. (2014) were not statistically significantly different in residential, institutional, or retail buildings, despite indoor PM_{2.5} mass concentrations being 60% lower than outdoor concentrations.^{1,5} Average indoor/outdoor (I/O) ratios of ROS on PM_{2.5} were 0.8±0.75 (retail), 1.02±0.55 (institutional), and 1.22±0.85 (residential). Similar observations were made using real-time instrumentation when windows were open and closed in an unoccupied residence where ROS on PM was measured to be similar indoors and outdoors, regardless of whether windows were open or closed (I/O ratio of 0.9-1.2), while PM₁ I/O ratios were 0.7 and 0.4 when the windows were open or closed, respectively.⁴ In the same study, I/O ratios of gas+particle-phase ROS concentrations differed when windows were open (0.8) and closed (0.4-0.5). The observation that outdoor gas+particle-phase ROS concentrations were higher than indoor gas+particle-phase ROS concentrations coupled with little difference between indoor particle-phase and gas+particle-phase ROS concentrations lead the authors to suggest the building was a sink for gas-phase ROS. With additional measurements of air exchange rate and ozone, sulfate, and volatile organic compound concentrations, the study hypothesizes that heterogeneous surface chemistry is a source of ROS and surfaces can act as a reservoir of ROS that partitions to indoor particles as an explanation for particle-phase ROS I/O ratios being close to 1, while PM I/O ratios are much lower.

Total suspended particulate (TSP) samples collected in eight homes indicated elevated outdoor ROS on TSP concentrations (2.35±0.57 nmol/m³) compared to indoors (1.59±0.33 nmol/m³), a result that may be impacted by differential removal of coarse particles containing ROS during particle penetration into buildings.⁶ In the same study, indoor ozone and terpene concentrations were varied in a test home when outdoor ozone concentrations were either high (>40 ppb) or low (<40 ppb). When outdoor ozone concentrations were low, it was estimated that 34% of ROS on TSP was from outdoors for the low indoor ozone and terpene condition, and outdoor ROS on TSP reduced to contributing 16% of indoor ROS on TSP for the high indoor ozone and terpene condition, suggesting significant indoor sources of particle-bound ROS. Outdoor ROS on TSP was estimated to contribute 41-51% of the measured indoor ROS on TSP under high outdoor ozone conditions, and the transport of ROS precursors into buildings was suggested to significantly influence indoor generation of particle-bound ROS.⁶

Recent measurement and modeling studies suggest semi-volatile ROS species can accumulate on surfaces and partition to particles.^{1,4,21,22} A modeling study focused on semi-volatile organic peroxides suggests a substantial amount of ROS on particles can partition from surface films.¹ Another modeling study estimated that 91-96% of ozone and H₂O₂ are deposited onto surfaces in a typical residence.²¹

Measurements of ozonolysis of a film composed of a mixture of lipids representing skin lipids and cooking oils showed depletion of about half of the ozone reacting to form ROS within the first few hours of exposure, and about half of the formed ROS persisted on the surface film. ROS production continued during the 24 hours following ozonolysis, suggesting other mechanisms for ROS production in surface films, such as autoxidation.²³

Indoor Sources and Control of ROS and RNS

Besides outdoor ROS and RNS penetrating building envelopes and partitioning of ROS from indoor surfaces onto particles, other sources of indoor reactive species include combustion (e.g., incense, cigarettes),^{20,22,24} cooking,^{7,25} oxidation of gas-phase terpenes (e.g., from surface cleaning),^{13,26} nitrous acid (HONO) photolysis,^{11,13,14,27} electronic/additive air cleaners,^{13,19,28-31} disinfectant spraying/fogging,^{31,32} laser printers,³³ and potentially through germicidal UV photolysis of ozone and volatile organic compounds.^{34,35} Chamber studies demonstrate significant ROS production resulting from limonene ozonolysis, a common indoor reaction.³⁶ Additional sources and the relative importance of the above sources may be identified in the future.

Methods to reduce indoor concentrations of particle-bound ROS and RNS include filtration and reducing organic surface film formation and/or oxidation. Compared to operating a test house without a filter installed, ROS on PM_{2.5} was reduced by 82% when a MERV16 filter was installed.⁵ Reducing the concentration of ROS on surfaces formed through oxidation, such as by reducing indoor ozone concentrations, should result in reduced partitioning of such compounds to the particle-phase. Reduction, source control, or elimination of emissions from human activities, such as cooking, smoking, incense burning, surface cleaning, and laser printer use, should also reduce indoor reactive species concentrations. It is also important to carefully select air management technologies to ensure reactive species production is not significant or can be mitigated. Additional control methods for reactive species may be identified in the future.

What does this mean for ASHRAE?

ROS and RNS are contaminants of concern due to their impact on health and can react with more benign compounds to produce contaminants of concern, such as secondary organic aerosols (SOA).¹⁴ Engineering systems can reduce the contribution of reactive species within buildings, such as installation of high performance filtration removing ROS on PM_{2.5}. Since reactive species may be generated or partition from surfaces indoors, there may be additional engineering controls that can reduce source strengths of ROS and RNS, such as stove hood exhaust systems. ASHRAE should understand the risks associated with reactive species and effective engineering interventions. In the long term, ASHRAE may need to change its existing standards or adopt new standards to provide industry guidance on addressing ROS and RNS exposure indoors.

What Actions Should ASHRAE Consider?

- Technical committees should engage with indoor chemists to better understand sources and control approaches for ROS and RNS indoors.
- Reactive species generation should be included in testing and validation of certain air quality control methodologies, such as additive air cleaner technologies and germicidal UV.³⁴ Specific attention should be paid to the concentrations and species produced, as well as how the produced reactive species may be mitigated.
- Identify knowledge gaps and support research, including but not limited to:
 - Indoor air quality monitoring and engineering controls to reduce indoor generated NO_x.

- The measurement and modeling of ROS and RNS in particles and indoor surface films.
- Physicochemical modeling of ROS and RNS production and reaction with indoor gases and surfaces.
- Assess the effectiveness of gas- and particle-phase ROS and RNS removal approaches, including filtration and adsorption.

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