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The past, present and future of indoor air chemistry

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The past, present and future of indoor air chemistry

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"In developed countries, we spend 80-90% of our time indoors" is the opening sentence for most grant applications and publications in our field. But has this well-worn truism lost its impact? We know that the majority of our exposure to air pollution occurs indoors, so why has indoor air quality not received the attention it deserves and how do we as a community better communicate this message? Recently, there has been increasing interest from experts from a wide range of backgrounds, including outdoor air quality scientists. They have enhanced our community, sparking a rapid evolution in the measurement technology used indoors, and the number, diversity and novelty of findings.

The INDoor AIR POLLution NETwork (INDAIRPOLLNET) was recently supported by the European Cooperation in Science and Technology (COST), following submission of a proposal that began with the sentence highlighted above. It consists of ~200 participants from 38 countries, comprising both scientists and practitioners in chemistry, biology, aerosol characterization, toxicology, exposure, emissions and chemical risk assessments, material design, building physics, civil engineering and standardization. Over a four-year period, INDAIRPOLLNET will address the current state of indoor air pollution, with emphasis on indoor air chemistry (IAC), including the associated research needs, challenges, and ways to address them. A liaison with the International Organization for Standardization (ISO) will facilitate the transfer of this scientific knowledge to practice.

Increasing climate change awareness is driving rigorous energy efficiency measures with buildings becoming more airtight, though adverse health effects can be associated with lower ventilation rates.[1] Air pollutant concentrations are often higher indoors than outdoors, particularly following activities such as cleaning, cooking and smoking.[2] More than two million healthy life years are lost across

Europe because of indoor air pollution, including indoor exposure to outdoor pollutants, indoor combustion sources, moisture, and emissions from building materials and consumer products.[3] There are vast differences in building types and uses, occupant behavior, geographical locations, ventilation systems and indoor and outdoor sources. Chemical processes indoors and their relation to those occurring outdoors must be well understood, in order to extrapolate results to a wider range of buildings and locations than in the relatively few, in which measurements have been made.

In its first year, INDAIRPOLLNET used seven subgroups to mine recent literature to summarize what existing measurement and model studies reveal about IAC. The research priorities from the subgroups are now presented.

Chemical transformations

Indoor chemical processes have been identified by reviewing laboratory, field and modelling studies both indoors and, where relevant, outdoors. The relative importance of chemical processes indoors differs to outdoors because of higher surface-to-volume ratios, and differences in pollutant sources and dispersion, light characteristics and temperature and humidity profiles.[4,5] The following priorities were identified:

- Assess the potential for indoor chemistry in the gas phase, with emphasis on:
  - Role of photolysis (via attenuated solar and artificial light sources),
  - comparison of the relative importance of OH, Cl, NO_3, O_3 as oxidants, the conditions where each dominates and routes to formation,
  - contribution of different reaction pathways to the formation of secondary products,
  - competition between chemical reactions and ventilation rate.
- Advance knowledge on the abundance of organic trace constituents in indoor air and the identification of new constituents through total OH reactivity measurements compared with measurable individual trace gas measurements.
- Investigate the impact of solid and liquid phase processes indoors (e.g. reactivity in the liquid phase indoors, including acid-base chemistry).
- Parameterize heterogeneous processes (production yields of secondary species such as aldehydes and HONO) for representative indoor surfaces and in real environments.

Building materials, household products and occupant behavior

Building and household products (e.g. wood based building materials, paints, varnishes, cleaning and personal care products, air fresheners, combustion appliances, electronic appliances, furniture, carpets, toys) as well as occupant activities (e.g. cooking, smoking, cleaning) can contribute significantly to indoor air pollution. A comparison of 13 labelling schemes for construction products worldwide has identified 15 lists of target compounds, with 611 individual chemicals occurring on at least one of the lists.[6] Indoor surfaces may act as both sources and sinks of gas-phase air pollutants; there is increasing interest in secondary pollutant emissions following surface interactions indoors.[5,7] The following are the identified research priorities:

- Improve the characterization of pollutant emissions, deposition and chemical transformations on various indoor surfaces for the identification and development of building and furnishing materials for better indoor air quality.
- Design field studies with a particular focus on the role of semivolatile organic compounds (SVOCs) and their reaction products in surface chemistry and related health effects.
- Disentangle the role played by humidity (potential impact on aqueous chemistry) versus reactive species (e.g. chlorine) in surface reactivity.

- Study the impact of building construction, location and operation on IAC.

Occupants

Humans emit a range of organic compounds from sweat and sebaceous secretions from skin, breath, and intestinal gases. The compounds associated with the presence of humans contribute 40-57% of the volatile organic compound (VOC) concentrations indoors in daytime.[8,9] Studies on occupant-related chemical transformations have mainly focused on ozonolysis of squalene, a major skin oil constituent.[10] The following research priorities were identified:

- Investigate the personal and environmental factors that influence human emissions (e.g. diet, stress level, personal hygiene, age, sex, health condition, activity level, personal care products, clothing and its laundering).
- Study the inter- and intrapersonal variability and the influence of prior exposure on human dermal and oral emissions and their reactive capacity.
- Perform real-time measurements of OH, NO_3, Criegee intermediates and other short-lived, highly reactive species in occupied and unoccupied indoor environments.
- Investigate the influence of occupancy on indoor surfaces as well as on the composition of airborne particles.

Microbial activity

Microbial life is ubiquitous within buildings[11] and is a frequent cause of indoor air quality problems and health effects.[12] Microbial colonies such as molds produce a wide variety of VOCs through their metabolisms, which encompass a range of functional moieties.[13] The behavior of analogous microbes in the ambient atmosphere suggests that their indoor counterparts are chemically and photochemically active and are likely to affect chemistries both in the gas phase and on surfaces, including on aerosols. Future research priorities include:

- Determine the relative VOC load from microbial activity and the impact on IAC in an "ordinary dwelling" compared to a "problem dwelling" with similar occupancy.
- Determine the influence of species variety indoors and of the balance between the building biome and the occupant biome on indoor air chemistry.
- Investigate the levels of toxic volatile emissions resulting from microbial processing of widely-used building materials.
- Coordinate a network of well-characterized test facilities with a minimum set of controlled variables in conjunction with a coupled indoor chemistry/dynamics model framework, in order to address the potential variability due to geographic differences in species, indoor environments and emission factors.
- Individually study microbial activity by the use of standardized coupons whereby known quantities of microbes are introduced into well-characterized realistic environments and the marginal effects of the microbial activity is measured.

Particles

Airborne particles form an integral part of IAC, as dynamic changes between gas and particle phase take place continuously.[14] Particles in indoor air are influenced by both physical and chemical processes, which change their physical characteristics, chemical composition and concentrations.[15] Recent studies based on real time aerosol mass spectrometry have brought novel understanding of chemical transformations taking place indoors. Future studies are recommended in the following domains:

- Study the factors influencing SVOC uptake on particles (e.g. chemical composition, number size distribution, surface area, environmental parameters).
- Determine size resolved particle- and gas phase chemical emission factors and mass spectra/signatures for specific indoor sources under controlled laboratory settings with parallel use of proton-transfer-reaction mass spectrometry and aerosol mass spectrometry.
- Assess the oxidative reactivity of particles from specific sources and mixtures under laboratory conditions and compare with measurements in occupied real indoor environments.
- Study the change of physicochemical characteristics of particles upon infiltration and the transformations that occur when outdoor and indoor air pollutants in the gas- and particle-phase interact.

Source apportionment

Receptor models apportion the measured mass of an atmospheric pollutant at a given site (receptor), to its emission sources by using multivariate analysis to solve a mass balance equation.[16] Positive Matrix Factorization (PMF), Chemical Mass Balance (CMB) and Principal Component Analysis (PCA) are among the receptor models most frequently used. While the major sources of indoor air pollution have been identified, few studies have attempted to estimate the contribution of specific sources using such techniques, mainly because the presence of both indoor and outdoor sources, building-related mechanisms (e.g. ventilation, infiltration), as well as outdoor meteorology and long-range transport of pollutants makes source apportionment challenging. Key research areas that warrant attention are:

- The influence of IAC on source apportionment model applications, including the validity of assumptions and relevant constraints (e.g. unstable source profiles over time).
- The dependence of receptor modeling applications on available decay rates, air exchange rates and penetration factors.
- Improved estimates of the outdoor contribution in source apportionment of indoor pollutants.
- Perform source apportionment studies based on datasets obtained with real time measurements with very short time resolution (e.g. aerosol mass spectrometer, PTR-MS).
- Define basic guidelines for source apportionment in indoor environments and address reliability issues of online source apportionments based on low-cost sensor networks.

Modelling

Indoor air pollutant measurement techniques are still unable to measure many pollutants at sufficient temporal frequency and with the required specificity in a wide enough range of buildings, to provide a broad and representative understanding of chemistry indoors. The development and use of indoor air models is, therefore, a substantial requirement for understanding IAC.[17] Indoor air chemistry models need to include the important sources and sinks of pollutants within a building envelope, such as chemical reactions, material emissions and surface interactions, human activity, exchange of pollutants with outdoors, or transport of pollutants within/between different zones of a building. A number of challenges for modelling studies have been identified:

- Field experiments in real buildings, especially those with real-time measurements and real/simulated activities, are necessary to validate and improve models.
- Models often assume well-mixed air in buildings, when spatial variation within zones should be considered.
- Indoor air chemistry models typically use chemical mechanisms originally constructed for modelling outdoor chemistry and may lack appropriate degradation schemes.

- Current models typically include estimated photolysis rates indoors (although some measurements have become available recently[18,19]) and often fail to consider the propagation of light from the windows throughout the indoor space.
- Modelers and experimentalists must work together; models help design experiments and the experimental results can be used to improve models.

The field of indoor air chemistry is moving forward rapidly, accelerated to a great extent by the Alfred P. Sloan Foundation's *Chemistry of Indoor Environment* program (whose website's homepage vividly flashes the "90%-sentence"). From the lab to the field, from test houses to climate chambers, from extraordinary campaigns such as HOMEChem[20] to modeling efforts like the international MOCCIE consortium, invaluable data about indoor air chemistry and physics is being swiftly generated. Indoor air chemistry occupies an increasing share in the programs of the Indoor Air conference series and at meetings such as the recent joint conference of The International Societies of Exposure Science (ISES) and Indoor Air Quality and Climate (ISIAQ) in Kaunas, Lithuania. But as it often is the case in science, new answers generate new questions and we seem to have lots of them. Such new questions are to be welcomed. As Albert Einstein noted, "To raise new questions, new possibilities, to regard old problems from a new angle, requires creative imagination and marks real advance in science". As we continue to address the unknowns of indoor air chemistry and allow our scientific curiosity to generate further insights, we should remember that we ultimately strive not only for understanding, but especially for healthier indoor environments.

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