

Conclusions

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Chapter 9 Conclusions

Jean-François Doussin

In the 1986 edition of their famous monograph, Finlayson-Pitts and Pitts defined experimental atmospheric simulation as "*Perhaps the most direct experimental means of examining the relationship between emissions and air quality*". This statement was, at the time, strongly supported by the enormous amount of research that had been conducted in the laboratory on chemical transformations of pollutants since the early work of Haagen-Smit et al. in the 1950s (Haagen-Smit et al. 1953; Haagen-Smit and Fox 1953).

Some of the early chamber studies, focusing on the interconversion of NO_x in the presence of VOCs and light, made a major contribution to the discovery of the role of the OH radical in the atmospheric photo-oxidation cycle (Heicklen et al. 1969, 1971). They also revealed the mechanism of tropospheric ozone build-up (Weinstock 1971; Niki et al. 1972; Westberg et al. 1971). The indefatigable work of Pitts, Winer, Atkinson, Niki, Becker, Moortgat, Schurath, and others led to the development of a huge database of kinetic reaction parameters that laid the foundations for the first chemical transport models. Subsequently, the use of simulation chambers has led to many other significant breakthroughs in atmospheric chemistry research. Simulation chamber experiments on the atmospheric oxidation of unsaturated hydrocarbons led to the identification of the essential role of biogenic VOCs in rural ozone formation (Abelson 1988). During the 1990s, chamber experiments contributed to the elucidation of the relation between gasoline composition and secondary organic aerosol formation (Odum et al. 1997), and more recently they were key in characterizing the major oxidation routes for organic aerosol in the atmosphere (Jimenez et al. 2009). By revealing that oligomerization processes were occurring in organic aerosol (Kalberer et al. 2006), chamber experiments provided a basis for questioning one of the most established schemes—the assumption that the molecular carbon chains of organic

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pollutants tend to fragment until ultimately $CO₂$ formation predominantly occurs during atmospheric oxidation.

Over the past few decades, progress in our understanding of the atmospheric oxidation of isoprene—one of the most important biogenic VOCs—illustrates the need for chamber experiments and also epitomizes the synergies between laboratory and field studies. Following the identification of molecular tracers in field samples (Claeys et al. 2004), simulation chamber studies demonstrated that isoprene, C_5H_8 , despite only having five carbon atoms, could be oxidized in the atmosphere to form SOA (Kroll et al. 2005).

Soon after, innovative work conducted in the SAPHIR chamber, using emissions from real plants as precursors (Kiendler-Scharr et al. 2009), demonstrated the ability of isoprene to scavenge OH from the forest atmosphere, redirecting the chemistry toward the formation of products less prone to participate in nucleation and almost suppressing new particle formation events. Later on, focusing on gas-phase processes and their consequences for the tropospheric radical budget, Fuchs et al. (2013) detected significantly higher concentrations of hydroxyl radicals than expected based on model calculations, providing direct evidence for a strong hydroxyl radical enhancement due to additional recycling of radicals in the presence of isoprene.

More recently, McFiggans et al. (2019) showed that isoprene can decrease the overall mass yield derived from monoterpenes in mixtures through the scavenging of highly oxygenated monoterpene products by isoprene-derived peroxy radicals. With this discovery, these authors did not only bring important pieces of observation, but they also questioned the additivity of aerosol yields implemented in models. They illustrated that modest aerosol yield compounds are not necessarily net producers and that their oxidation can suppress both particle number and mass for stronger contributors present in the same air mass.

Around the same time as the comments from Finlayson-Pitts and Pitts in the late (1980), the International Union of Pure and Applied Chemistry (IUPAC) set about defining "Smog Chamber in atmospheric chemistry" as follows (Calvert 1990): "*A large confined volume in which sunlight or simulated sunlight is allowed to irradiate air mixtures of atmospheric trace gases (hydrocarbons, nitrogen oxides, sulfur dioxide,* etc*.) which undergo oxidation. In theory these chambers allow the controlled study of complex reactions which occur in the atmosphere. However, ill-defined wall reactions which generate some molecular and radical species (e.g. HONO, CH2O, OH-radicals,* etc*.) and remove certain products (H2O2, HNO3,* etc*.), the use of reactant concentrations well above those in the atmosphere, ill-defined light intensities and wavelength distribution within the chamber, and other factors peculiar to chamber experiments require that caution be exercised in the extrapolation of results obtained from them to atmospheric system*".

This statement may appear to be rather pessimistic when considering the significant progress in atmospheric chemistry that chambers have facilitated. Nevertheless, each of the reservations expressed by IUPAC is not without relevance and the international chamber community has worked hard to address these challenges. In Europe,

a coordinated approach has been adopted through the various EUROCHAMP initiatives, which have further investigated way to improve the robustness of experimental simulation results. Even if the 1990 IUPAC definition for "Smog Chamber" was still in use in the last edition of the IUPAC "Gold Book" (Chalk 2019), the readers of the present guide to atmospheric simulation chambers would be able to recognize the considerable advances that have been made in the field over the last 30 years.

9.1 Improving the Robustness of the Simulation Chamber Experiments

For many years, simulation chamber studies were conducted using reactant concentrations several orders of magnitude larger (ppm or hundreds of ppb levels) than those found in the ambient atmosphere (ppb or sub-ppb). However, thanks to the development of more and more sensitive monitoring techniques, working in the ppb range is now relatively standard and the ppt range is also accessible, although still challenging due to sensitivity limitations of measurement techniques. For decades, working with reactant concentrations well above those found in the atmosphere was not considered as a major problem, as long as non-atmospherically relevant radical–radical reactions were kept negligible. However, the highly non-linear nature of secondary aerosol formation and related condensation processes made it more critical to work at realistic concentrations. The conceptual advance brought by Pankow (1994) and Odum et al. (1997) has partially allowed us to take this common drawback of simulation chamber experiments into account when deriving SOA yields. Nevertheless, it was quickly shown (Duplissy et al. 2008) that the chemical composition of the organic aerosol formed, and therefore its physical properties, such as hygroscopicity and CCN activity, depended on the initial concentration of the precursors. Fortunately, demonstration of the critical need for reducing reactant concentrations in chamber experiments arrived around the same time as the introduction of a new generation of very sensitive mass spectrometry techniques such PTR-MS, API-TOF, TOF-CIMS,… which are providing further opportunities for simulation chamber studies to be performed at realistic atmospheric concentrations.

Significant progress has also been made in the characterization of light intensity inside the chamber (including homogeneity) and the provision of a wider range of wavelengths for simulating sunlight-induced atmospheric processes (Chap. 2). Even though UV fluorescent tubes, or so-called "black lights", are still—for cost reasons—the most common light source among the simulation chamber community, and even if the atmospheric relevance of their emission spectrum can be questioned, the recording of related actinic flux information together with chamber data has become a well-accepted practice, thanks to projects such as EUROCHAMP. This good practice is further supported by the information provided in Chap. 2 of this book, which provides a solid basis for homogeneous robust lighting characterization. The

availability of this information is indeed critical for any attempt of re-using previous datasets, especially if a modeling approach is planned (which is generally the case).

One of the key steps in the long journey of the scientific community toward a precise understanding of the chemistry at work in atmospheric simulation chambers is the rise of the concept of "chamber chemistry". According to this concept, when the initial precursor concentration is low enough and when the light energy is atmospherically relevant, the observed behavior of chemicals during chamber experiments would be the result of the interplay between the atmospherically relevant chemistry and the chamber effects.

Refusing to consider the chambers as black boxes and hence starting to study the chamber-dependent processes themselves with scientific rigor has led to two complementary efforts. The first of these is the study of wall reactions using the tools of microphysics and surface chemistry. While some aspects of wall reactions are relevant for the understanding of atmospheric processes (see, e.g., Pitts et al. 1984; Rohrer et al. 2005), the true motivation for characterizing them was a different one: the authors were already building what is now known as an "auxiliary mechanism". Thirty years ago, Jeffries et al. (1992) were already recommending that those chamber-dependent reaction sets should be available for each chamber dataset to be simulated. They were pointing out that when evaluating a reaction mechanism in a given chamber, the auxiliary mechanism is combined with a core mechanism which is asserted to be chamber independent, and that misrepresentations in the auxiliary mechanism could induce compensating errors in the core mechanism and so in the derived atmospherically relevant knowledge. This goal has never been that close to being attained, as not only this information has been made available for most of the atmospheric simulation chambers installed in Europe, but also because the present guide, for the first time, provides clear guidance for the building of such auxiliary mechanism (see Chaps. 2 and 3).

The second aspect of chamber-dependent processes is related to the simultaneous exploitation of a large number of datasets arising from various chambers. Based on a quasi-statistic interpretation of the previous concept, it assumes that deconvolution of chamber-dependent processes from atmospherically relevant processes can be significantly enhanced by the parallel analysis of comparable experiments carried out in different simulation chambers. This approach has been proposed as early as in 1999 (Jeffries 1999), but until now it has never been applicable due to the lack of diversity among datasets. In most of the centers developing an experimental atmospheric simulation activity, datasets are indeed carefully stored, forming several databases comprising generally hundreds to thousands of experiments. Nevertheless, the community was missing coherent datasets investigating the same chemical systems by means of very different installations. Thanks to the "multi-chamber experiments" initiative developed in the framework of EUROCHAMP-2020, this approach for three important "standard" chemical systems (propene oxidation, toluene/xylene oxidation, and a-pinene oxidation) may receive its first full-scale validation. Further, the release of the EUROCHAMP database (<https://data.eurochamp.org/>) has made freely available around 3000 datasets of chamber experiments, generated in more

than 20 fully characterized chambers (including an auxiliary mechanism for 16 of them), which exhibit a high diversity of size, type, material, and irradiation.

Together with the present guide, the EUROCHAMP database provides an unparalleled opportunity to ascertain a deeper understanding of these experiments and their implications for atmospheric processes, air quality, and climate. The combined use of data mining and emerging artificial intelligence techniques may further help to stimulate movement toward a thorough reanalysis of experiments in the database.

9.2 Simulating the Complexity of the Real Atmosphere, Working at the Interfaces and Considering Longer Timescale Exposure

For decades, the usual way of operating chambers has been to study a well defined but simplistic starting mixture with the goal of understanding all the mechanistic details of the transformation at work. This approach is still very valuable for characterizing the atmospheric processing and impact of a single compound or to study a well-defined chemical reaction. Leaving aside successive improvements made to the chamber experiments which adopt this "classical" approach to atmospheric simulation, ongoing advances continue to include a resolute movement toward more complex mixtures and more realistic systems, some of which may include including several phases of matter or interactions of chemicals with various surfaces.

From studies of the chemical evolution of emissions from real plants (Joutsensaari et al. 2005; Mentel et al. 2009; Faiola et al. 2018), motor vehicles (Geiger et al. 2002; Platt et al. 2013; Gordon et al. 2014), and wood burning devices (Nordin et al. 2015; Pratap et al. 2019), most of the emitting systems or practices utilize simulations in the attempt to characterize their impact on secondary atmospheric pollution, to evidence interplay between intermediate species arising from various precursors, or to identify tracers for their specific contamination.

Even more challenging is the application of simulation chambers to the interfaces between Earth system compartments. As shown in Chaps. 7 and 8, work on interfacial processes that was historically focused on the gas–aerosol or gas–liquid interfaces is now being extended to air–urban surfaces (Monge et al. 2010), air–sea exchanges (Bernard et al. 2016), as well as interfaces in the cryosphere (Thomas et al. 2021). In this case, not only the complexity of chemical mixture has increased, but the involvement of new surfaces for accommodation and for reaction implies to consider transport to/toward new media and complex mass transfer between phases.

Finally, a third dimension of complexity has recently emerged extending the timescale of simulation experiments. Most of the effects of modern air pollution on health, plants, or cultural heritage are related to long-term exposure: after decades of studies using exposition chambers in which primary pollutants are injected, simulation chambers are now considered as tools of choice to investigate the effect of complex mixtures containing secondary pollutants. For health impact studies, even if very sensitive models (such as pregnant mice or bare epithelial cells) are sometime used, it remains necessary to expose them to simulated polluted air for several days or weeks when the longest experiments in chambers generally last up to 2 to 3 days. To overcome these limits, new protocols are emerging, where chambers are generally operated as slow flow reactors with a constant input of primary pollutants that are allowed to react for an average time equal to the residence time (generally a few hours). When a steady state is attained, such a design can be operated for days providing a steady system (which limits the study of photochemical processes to indoor chambers) and feeds exposition devices where living models are receiving chamber effluents.

This area of research is ongoing and the protocols are still under development but they already benefit from those described in the present guide. Undoubtedly, the present effort in disseminating good practices and harmonizing protocols and the related metadata will have to be continued in the near future. It is probably one of the most critical networking activities that will have to be organized within the ACTRIS European Research Infrastructure.

9.3 Conclusion

The original use of smog chambers for the understanding of chemical transformations in the atmosphere; for the quantification of reaction rates, the extent, and the relevance of various possible pathways; and for the identification of secondary pollutants is still strongly necessary. The models—both operational and research oriented—are still far from an explicit thorough inclusion of all the processes that are required to represent and forecast the actual air quality and climate issues as well as future challenges. At the same time, the field of atmospheric experimental simulation has been extremely active during the past 15 years—and considering the number of new facilities around the world—there is little doubt about its vitality over the next 15 years, and beyond. A number of new methodologies and applications have risen and they will bring the operational capacity of simulation chambers to a new level. This community effort will enable a much broader range of scientific and societal challenges to be addressed, including not only the direct and indirect climate effects of atmospheric pollutants, but also the impact of air composition on health, cultural heritage, and the various compartments of Earth system. Although these applications are still in their early stages, they are quickly growing and are already producing data that will open new ways to consider the interplays between atmospheric transformations and impacts.

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