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Chapter 1

Introduction to Atmospheric Simulation Chambers and Their Applications



Astrid Kiendler-Scharr, Karl-Heinz Becker, Jean-François Doussin, Hendrik Fuchs, Paul Seakins, John Wenger, and Peter Wiesen

Abstract Atmospheric simulation chambers have been deployed with various research goals for more than 80 years. In this chapter, an overview of the various applications, including emerging new applications, is given. The chapter starts with a brief historical overview of atmospheric simulation chambers. It also provides an overview of how simulation chambers complement field observations and more classical laboratory experiments. The chapter is concluded with an introduction to the different aspects requiring consideration when designing an atmospheric simulation chamber.

Atmospheric simulation chambers, such as those in the EUROCHAMP network, are highly valuable research tools for investigating chemical and physical processes that occur in air. They are used in a large number of applications, ranging from air quality and climate change to cloud microphysics, cultural heritage and human health. Chambers were originally developed as laboratory-based systems to investigate the formation of clouds or photochemical smog and hence, were called cloud chambers or smog chambers, respectively. Their ability to provide a controlled environment to study the formation and evolution of atmospheric pollutants, by isolating

Astrid Kiendler-Scharr passed away before publication of this chapter.

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specific compounds of interest and controlling the oxidizing environment, made them especially useful in elucidating the key factors governing photochemical smog formation on a local to regional scale. Within EUROCHAMP-2020 and across the world, chambers dedicated to the exploration of atmospheric chemistry outnumber the atmospheric physics and cloud chambers. For this reason, this guide has an emphasis on atmospheric chemistry related aspects of simulation chambers.

Initially, smog chamber experiments were focused on elucidating the processes responsible for the observed increase in atmospheric secondary pollutants such as ozone and peroxyacyl nitrates (PAN-type compounds). This approach was later broadened to include studies of the kinetics and mechanisms of gas phase atmospheric oxidation and chambers have been extremely useful in producing kinetic data, branching ratio and product distributions (Becker 2006). Together with data arising from flow tubes and flash photolysis experiments, this knowledge allowed the scientific community to build complex numerical chemical codes that have led to the development of the models used to predict ozone formation. Nowadays, chambers are also essential tools for evaluating these chemistry models and for predicting the formation of secondary pollutants in the absence of uncertainties associated with emissions, meteorology and mixing effects (Carter and Lurmann 1991; Dodge 2000; Hynes et al. 2005). Experimental chamber data have been key to the development and optimisation (e.g. Gery et al. 1989; Carter 2010; Bloss et al. 2005a), as well as the evaluation (e.g. Saunders et al. 2003; Goliff et al. 2013; Jenkin et al. 2012; Bloss et al. 2005b; Metzger et al. 2008; McVay et al. 2016) of chemical mechanisms used in a wide range of science and air quality policy models. Today, chamber-derived data remains a key component in the development and evaluation of future atmospheric chemical mechanisms (Kaduwela et al. 2015; Stockwell et al. 2020).

In the past few decades, chamber facilities have been increasingly used to investigate processes leading to secondary organic aerosol (SOA), an important component of atmospheric aerosol (Finlayson-Pitts and Pitts 1986; Dodge 2000; Finlayson-Pitts and Pitts 2000; Kanakidou et al. 2005; Barnes and Rudzinski 2006; Hallquist et al. 2009). The general methodology which has been (and still is) useful for gaseous pollutants is now providing valuable data related to SOA formation (e.g. Hatakeyama et al. 2002; Pankow 1994; Odum et al. 1996; Cocker et al. 2001; Pun et al. 2003; Takekawa et al. 2003; Martin-Reviejo and Wirtz 2005; Baltensperger et al. 2005; Donahue et al. 2005; Pathak et al. 2007; McFiggans et al. 2019; Zhao et al. 2018, Ciarelli et al. 2017) as well as the physico-chemical properties of aerosols and their changes during atmospheric transport and processing (De Haan et al. 1999; Kalberer et al. 2006; Field et al. 2006; Linke et al. 2006; Meyer et al. 2009; D'Ambro et al. 2017; Huang et al. 2018; Zhao et al. 2017).

Furthermore, due to the wide range of experimental requirements, simulation chamber designs vary considerably. As pointed out by Finlayson-Pitts and Pitts (2000), although the general aims of all chamber studies are similar—i.e. to simulate processes in ambient air under controlled conditions—the chamber designs and capabilities to meet these goals vary widely. This in turn means that chambers and their associated measurement technologies are being adapted to a growing number of applications.

This chapter provides a short history of atmospheric simulation chambers Sect. 1.1, investigations of atmospheric processes Sect. 1.2, approaches for bridging the gap between laboratory and field studies Sect. 1.3, emerging new applications Sect. 1.4, and considerations on the design and instrumentation of atmospheric simulation chambers Sect. 1.5. Respective references to the more detailed discussion in Chaps. 2–8 are provided in each of the subsections.

1.1 A Short History of Atmospheric Simulation Chambers

Atmospheric simulation chambers have been used for more than 80 years. As early as the 1930s, Findeisen performed studies on cloud droplet size distributions and conducted cloud chamber experiments, which was a highly novel approach at the time. Findeisen's cloud chamber was approximately 2 m³ in volume and connected to a vacuum pump, which allowed the process of adiabatic expansion and atmospheric cloud formation to be mimicked in the chamber (Storelvmo and Tan 2015).

Photochemical smog formation, first observed in the Los Angeles area in the 1940s and 1950s stimulated study in large chambers to simulate plant damage and health effects such as eye and lung irritation (Haagen-Smit 1952). Europe followed suit in chamber construction and application to atmospheric processes and through a range of national and European Union funding streams, Europe now leads the world in the use of large, highly instrumented chambers for atmospheric model development and evaluation. These large facilities are complemented by a range of smaller chambers that have been designed for specific purposes.

The first large European chamber was the “Große Bonner Kugel” (Groth et al. 1972), constructed at the University of Bonn and completed in 1968. The programme led by Groth and Harteck initially focused on air glow reactions at the low pressures pertaining to the *upper* atmosphere. However, studies of tropospheric interest were also undertaken, but at a very basic level and without the use of photolytic sources. Radicals were generated by discharge flow techniques, and this limited the range of conditions that could be used.

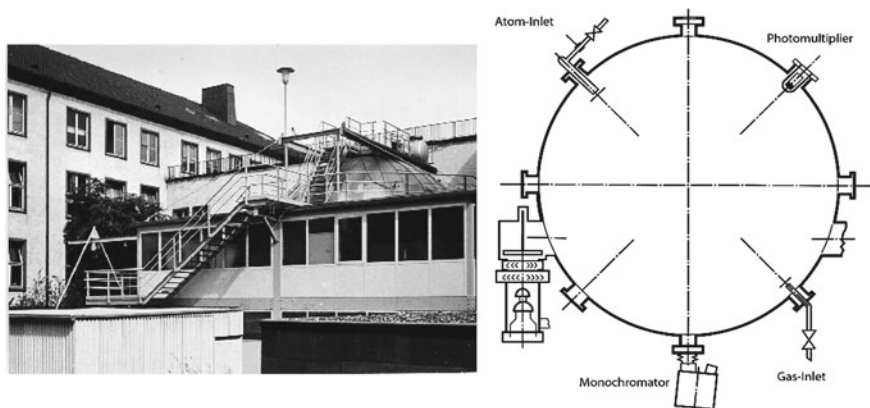
The facility, which was operated by Becker, Fink, Kley and Schurath for several years (Groth et al. 1972), had the following properties as indicated in Table 1.1.

At that time dark OH radical sources and the importance of OH reactions were not known. Figures 1.1 and 1.2 show the facility installed at the Institute of Physical Chemistry, Bonn University. The chamber has not been used since the mid-1980s because of its enormous operational cost and has since been completely dismantled.

In the mid-1970s, as our understanding of the basics of tropospheric chemistry increased and particularly the role of photolysis, the Pitts group at Riverside (Finlayson-Pitts and Pitts 1986, 2000) started to construct an indoor chamber with the objective of exploring photochemical smog formation. Advances in the understanding of photochemical processes had been slow because appropriate analytical techniques still had to be developed at that time. However, activity soon increased with the construction of a similar chamber in Japan (Akimoto et al. 1979a, b), while

Table 1.1 Key properties of the “Bonner Kugel”

Volume	221 m ³
Surface	177 m ²
Inner diameter	7.5 m
Heatable	$T_{\max} = 350\text{ °C}$ with 233 kW power, cooling of baffles between pumps and chamber with liquid hydrogen
Material	Stainless steel, 10 mm wall thickness
Pumping speed	240 000 l/s with 8 diffusion pumps
Lowest pressure	10 ⁻¹² bar

**Fig. 1.1** The housing of the “Große Bonner Kugel” (left) and cross-section through the spherical reaction chamber “Große Bonner Kugel” (right). Courtesy of K.H. Becker, Bonn, Germany

Atkinson in the Pitts group started to successfully investigate the kinetics of the initiation reactions of OH, O₃ and NO₃ with volatile organic compounds (VOC). Concurrently, other groups used Teflon bags to study smog-forming reactions under irradiation by natural sunlight, but their results were limited to the Los Angeles conditions.

The importance of the OH radical in atmospheric chemistry had been promoted by Weinstock (1969), working at the Ford Motor Company research laboratories at Dearborn. In this laboratory, Niki used a relatively small photoreactor to develop the application of FTIR spectroscopy for quantitative investigation of atmospheric reactions (Niki et al. 1972, 1981; Wu et al. 1976). IR absorption spectroscopy had been used for a number of years to study atmospherically relevant chemical reactions (Stephens 1958; Hanst 1971), based mainly on mirror systems which allowed long path light absorption (White 1942, 1976; Herriott et al. 1964; Herriott and Schulte 1965). However, it was the use of FTIR methods by Niki et al. (1981) and additional work in the Pitts’ group to quantitatively measure rate coefficients and products in photoreactors by long path FTIR absorption spectroscopy that really accelerated and

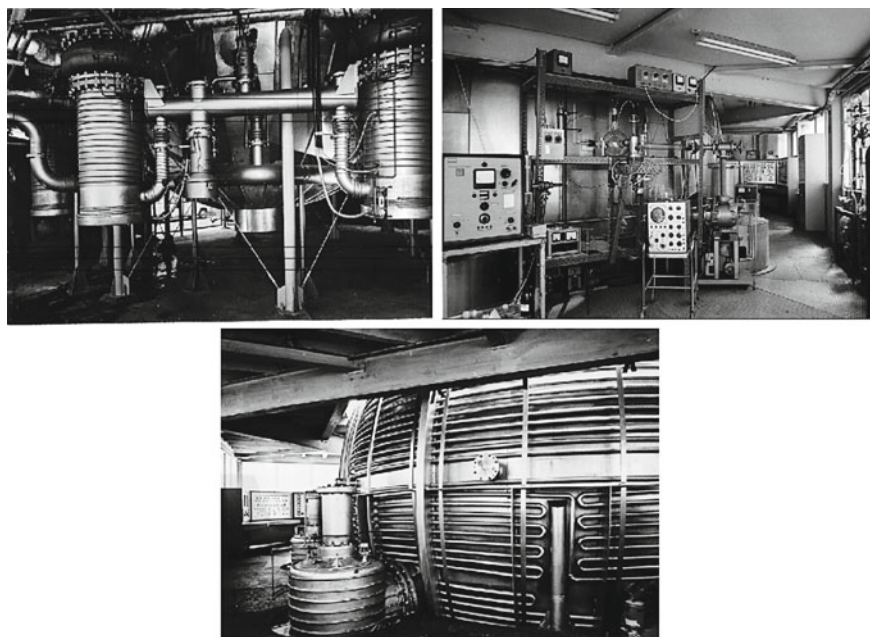


Fig. 1.2 The pipe system by which the chamber could be heated to 300 °C (left), enormous pumping capacity needed to reach the vacuum of 10⁻⁹ Torr (middle), the platform at which the experiments were prepared and carried out (right). Courtesy of K.H. Becker, Bonn, Germany

promoted the use of the technique and FTIR has been one of the work-horses of chemical simulation chambers ever since.

In the 1960s and '70s, the understanding of atmospheric reactions developed as first the key role of the OH radical was recognised as the dominant oxidizing agent in the troposphere, based on the analysis of the CO budget (Heicklen et al. 1969; Weinstock 1969; Stedman et al. 1970; Levy 1971), and the measurement of the OH + CO rate coefficient two years earlier (Greiner 1967). The propagation of an OH radical chain was understood 10 years later when the rate coefficient of the fast reaction $\text{HO}_2 + \text{NO} \rightarrow \text{OH} + \text{NO}_2$ was measured by several groups (Howard and Evenson 1977; Leu 1979; Howard 1979, 1980; Glaschick-Schimpf et al. 1979; Hack et al. 1980; Thrush and Wilkinson 1981), initiated by studies of Crutzen and Howard (1978) that showed the importance of this reaction in stratospheric ozone chemistry.

In Europe in the 1970s, several groups e.g., Becker and co-workers in Bonn and Cox and co-workers in Harwell, started studies on tropospheric chemistry based on either the technique of long path FTIR absorption spectroscopy in simulation chambers by Becker and co-workers in Wuppertal or molecular modulation studies focusing more on elementary reactions by Cox. Becker and co-workers constructed a multiple reflection mirror system in a 420 L photoreactor, which could be operated between 223 and 323 K to determine the OH reaction rate coefficients in combination with product analyses in the ppm range. Subsequent developments involved the

construction of a 6 m long quartz glass reactor of 1000 L volume, the QUAREC chamber, which enabled measurements to be extended down to the ppbV level. Over the years, other European laboratories started to use indoor chambers of larger volume irradiated by a range of photolysis sources (Baltensperger et al., in Villigen/Zürich, Carlier and Doussin in Paris, Hjorth et al., in Ispra, Herrmann et al., in Leipzig, Le Bras et al., in Orléans, Treacy et al., in Dublin, Wenger et al., in Cork). Tables 1.2 and 1.3 lists the larger indoor and outdoor reactors, respectively, that have been built up to 2000.

Large outdoor simulation chambers have many advantages in terms of photochemical smog simulation and several large outdoor chambers have been built in the US, with support from the EPA. A major objective of these studies was to determine ozone formation isopleths under chemical conditions representative of conditions observed in major US cities. These chambers were made from FEP Teflon foil, with volumes up to 25 m³. Whilst they lead to improvements in the empirical understanding of smog formation, the results could not be generalised because of the limited range of conditions requested by the US EPA. In Riverside, Carter and co-workers developed a method to define the ozone formation potential of VOCs by determining maximum incremental reactivity (MIR) factors using chamber data and chemical modelling (Carter 1994). A similar method was introduced by Jeffries in Chapel Hill, who also used an outdoor chamber (Fox et al. 1975).

Other approaches involved the injection of real engine exhaust directly into a smog chamber and studying the formation of ozone. However, the data were still very US specific in terms of the VOC/NO_x ratios and so could not be generalised and applied in other countries. In parallel, with the simulation studies mentioned above, Atkinson and co-workers refined their method to determine the OH reactivity from relative rate measurements in chambers and developed structure reactivity relationships to calculate rate coefficients for OH radical reactions with VOCs (Atkinson 1986, 1987; Kwok and Atkinson 1995). Further developments in simulation work included work by Seinfeld and co-workers in the mid-1980s, to study secondary organic aerosol formation from the oxidation of aromatic and biogenic hydrocarbons via the use of a 65 m³ outdoor chamber made of FEP Teflon (Pandis et al. 1991).

In Europe, the first development of a large, highly instrumented chamber was led in the mid-1990s, by Becker, Millán and co-workers who built the EUPHORE (European Photoreactor) outdoor chamber in Valencia, Spain. In fact, EUPHORE consists of two chambers made of FEP Teflon foil, each of which has a volume of 200 m³ (Becker 1996). This facility became a centre for European laboratories to work co-operatively on mechanistic, kinetic and ozone formation studies using either controlled starting materials or real exhaust gases from gasoline and Diesel engines. The EUPHORE chambers were equipped with a comprehensive suite of analytical instrumentation, including in situ detection of the key radicals HO₂ and OH using laser-induced fluorescence measurements.

In 2000, the group of Wahner at Forschungszentrum Jülich, Germany, built a new double walled outdoor chamber called SAPHIR (Brauers et al. 2003), which has a volume of 280 m³, see Fig. 1.3. The double wall made of FEP Teflon foil allows studies of oxidation processes at low NO_x concentrations (below 1 ppbV). The Jülich

Table 1.2 Indoor chambers without light sources or irradiated by black lamps or solar simulators up to the year 2000

Year	References and location	Description	Application
1968	Groth et al. (1972) Bonn	Dark chamber 220 m ³ , stainless steel (high vacuum)	Without light sources, for low pressure studies
1972	Niki et al. (1972), Dearborn	150 l Pyrex	Kinetic and mechanistic studies
1975	e.g. Doyle et al. (1975), Riverside	6 m ³ , evacuable, thermostated, FEP ^a coated aluminium	Photooxidant and kinetic studies
1979	Akimoto et al. (1979a), Tsukuba	6 m ³ evacuable, thermostated, FEP coated aluminium	Photooxidant studies
1980	Winer et al. (1980), Los Angeles	6 m ³ evacuable, thermostated, FEP coated aluminium	Photooxidant studies
1981	Barnes et al. (1979), Wuppertal	420 l Duran glass, evacuable, thermostated – 50 to + 50 °C	Gas phase studies
1982	Joshi et al. (EPA), Research Triangle Park	440 glass reactor	Photooxidant studies
1986	Barnes et al., Wuppertal	Quartz glass 1100 l, evacuable, thermostated 0 to + 25 °C	Gas phase and aerosol kinetic and mechanistic studies
1986	Evans et al. (1986), Australia	4 × 200 l FEP bags	Photooxidants studies
1988	Behnke et al. (1988), Germany, Hannover, now Bayreuth	ca. 3000 l, Duran glass, thermostated –25 °C to ambient temperature	Aerosol studies
1997	Möhler et al. (2001), Karlsruhe	84 m ³ , thermostated –90 to + 60 °C, <i>AIDA</i>	For trace gas, aerosol and cloud studies
1996	Wahner et al. (1998), Jülich	256 m ³ , FEP wall cover of a lab room	Without light source, for NO _Y chemistry
1997	Doussin et al. (1997), Paris	977 l, glass	Gas phase mechanistic studies
1998	Cocker et al. (2001), Pasadena	2 × 28 m ³ , 10–40 °C	Aerosol studies
2000	Carter et al. (2005), Riverside	FEP, double wall	Low NO _x studies

^a FEP fluorinated ethylene propylene

Table 1.3 Outdoor chambers irradiated by sunlight up to the year 2000

Year	References and location	Volume, wall material
1976	Jeffries et al. (2013), Chapel Hill	25 m ³ , FEP
1981	Fitz et al. (1981)	40 m ³ , FEP
1983	Spicer (1983)	17.3 m ³ , FEP
1985	Kelly (1982)	450–2000 l, FEP bags
1985	Jeffries et al. (1976), Chapel Hill	25 m ³ , FEP
1985	Leone et al. (1985), Pasadena	65 m ³ , FEP
1995	Becker (1996), Valencia	2 × 200 m ³ , FEP, <i>EUPHORE</i>
2000	Wahner (2002), Jülich	270 m ³ , FEP, double wall, <i>SAPHIR</i>

group did pioneering work in field measurements of OH and HO₂ concentrations (Hofzumahaus et al. 2009), so SAPHIR is fully equipped with the most advanced in situ radical measurement techniques (Fuchs et al. 2012a, b). A smaller double wall indoor chamber was recently built by Carter in Riverside, to study tropospheric oxidation processes at low NO_x concentrations.

Two other chambers were built in Germany, at the same time, for the study of aerosol processes. In 1986, Zetzsch and co-workers built a 3000 l Duran glass indoor chamber in Hannover, covered inside with FEP, and irradiated by solar simulators. This facility has been moved to Bayreuth. In 1987, Schurath and co-workers started to

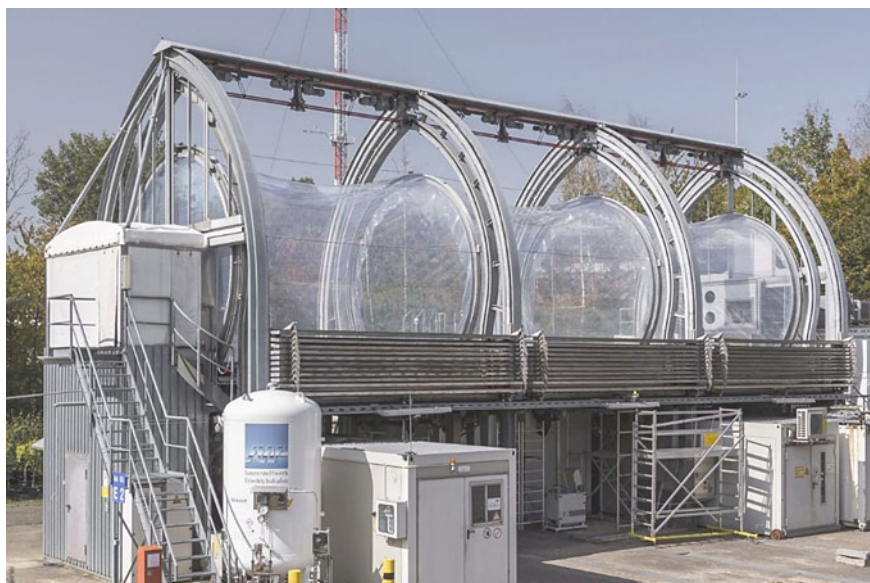


Fig. 1.3 The double wall outdoor chamber SAPHIR in Jülich, Germany (© “Forschungszentrum Jülich/Sascha Kreklau”)

operate the 84 m³ aluminium chamber AIDA (Aerosol Interaction and Dynamics in the Atmosphere) in Karlsruhe, which has homogeneous temperature control between +60 °C and −90 °C for trace gas, aerosol and cloud process studies. Other groups also now operate medium sized chambers.

A milestone for the European landscape of atmospheric simulation chambers was the implementation of the EUROCHAMP initiative, which started in May 2004 with the goal of joining together the existing European facilities into one integrated infrastructure of atmospheric simulation chambers.

The integration of all these chamber facilities within the framework of EUROCHAMP, followed by the EUROCHAMP-2 and EUROCHAMP-2020 projects, promoted the retention of Europe's international position of excellence in this area and it is unique in its kind worldwide. The mobilization of a large number of stakeholders dealing with environmental chamber techniques provided an infrastructure to the research community at a European level, which offers maximum support for a broad community of researchers from different disciplines. Overall, the EUROCHAMP projects fostered the structuring effect of atmospheric chemistry activities performed in European chambers and initiated wider international collaborations by supporting transnational access activities. Nowadays these facilities are fully available for the whole European scientific community and are exploratory platforms within the new Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS). The following tables summarize current chambers across the world (Table 1.4) starting with the chambers of the EUROCHAMP consortium.

1.2 Investigations of Atmospheric Processes

1.2.1 *Reaction Kinetics and Product Studies*

Being the building blocks of the general atmospheric chemical mechanism, the study of the kinetics of elementary steps and the related product distribution has been the main application of simulation chambers. Involving pure gas phase conditions this has been—and is still—often carried out in small photoreactors of a few hundred litres or in small indoor simulation chambers. In the case of kinetics studies, Teflon bags of several litres to a few cubic-meters working under atmospheric pressure and ambient temperature under artificial irradiation (generally UV fluorescent tube) were often used to apply relative rate methods (Brauers and Finlayson-Pitts 1997). Nevertheless, the atmospheric fate of hundreds of various volatile organic compounds (VOC) was also studied—and is still—in rigid chambers such as the one displayed in Fig. 1.4 (Barnes et al. 1987; Doussin et al. 1997; Etzkorn et al. 1999; Picquet-Varrault et al. 2001; Atkinson 2000). This systematic kinetic and mechanistic work has produced over time a comprehensive database that has established the foundations of most chemical schemes used in numerical models.

Table 1.4 List of current chambers across the world; The chambers of the EUROCHAMP consortium are listed in alphabetic order of chamber acronym, and chambers across the world outside the EUROCHAMP consortium are listed in alphabetic order of country

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
<i>EUROCHAMP chambers</i>							
AIDA KIT, Karlsruhe, Germany	Indoor	Aluminium	85	LED	+ 60 °C to – 90 °C	Evacuable and developed for aerosol and cloud simulation	Möhler et al. (2003), Wagner et al. (2006)
CESAM CNRS-UPEC, Créteil, France	Indoor	Stainless steel	4,2	Xenon arc lamps	– 10 to + 50 °C	Evacuable and developed multiphase atmospheric processes	Wang et al. (2011)
CHAMBRé INFN, Genoa, Italy	Indoor	Stainless steel	3	Arc	Ambient	Evacuable and suitable for bioaerosols	Massabò et al. (2018)
CSA CNRS-UPEC, Créteil, France	Indoor	Pyrex	0,97	Fluorescent Tubes	Ambient	Evacuable Gas phase chemistry	Doussin et al. (1997)
ESC-Q-UAIC CERNESIM, Iasi, Romania	Indoor	Quartz	0,76	Fluorescent Tubes	Ambient	Gas phase chemistry	
EUPHORE CEAM, Paterna, Spain	Outdoor	FEP	200	Sun	Ambient	Dual chambers gas phase and aerosol	Zádor et al. (2006)
FORTH-ASC FORTH, Patras, Greece	Indoor	FEP	10	Fluorescent tubes	Ambient	Indoor simulation chamber	Kostenidou et al. (2013), Kaltsounidis et al. (2017)
FORTH, Patras, Greece	Outdoor	FEP	2	Sun/Fluorescent tubes	Ambient	Mobile	Kaltsounidis et al. (2019)

(continued)

Table 1.4 (continued)

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
HELIOS CNRS_Orléans, Orléans, France	Outdoor	FEP	90	Sun	Ambient		Ren et al. (2017)
CNRS_Orléans, Orléans, France	Indoor	FEP	16	Fluorescent tubes	Ambient		Le Person et al. (2008)
HIRAC University of Leeds, Leeds, UK	Indoor	Stainless steel	2	Fluorescent tubes	-25 to +70 °C	Evacuable gas phase chemistry and radicals	Glowacki et al. (2007)
IASC UCC, Cork, Ireland	Indoor	FEP	27	Fluorescent tubes	15–25 °C	Atmospheric chemistry	
ILMARI University of Eastern Finland, Kuopio, Finland	Indoor	FEP	29	Fluorescent tubes	16–25 °C	Combustion emission and health	Leskinen et al. (2015)
ISAC CNRS-IrceLyon, Villeurbanne, France	Indoor	FEP	2	Fluorescent tubes	Ambient		Bernard et al. (2016)
ACD-C TROPOS, Leipzig, Germany	Indoor	FEP	19	Fluorescent tubes	Ambient	Dual chamber	

(continued)

Table 1.4 (continued)

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
MAC University of Manchester, Manchester, UK	Indoor	FEP	18	Xenon arc lamp	Ambient	Can be coupled with the MICC cloud chamber	Alfarra et al. (2012)
QUAREC Wuppertal University, Wuppertal, Germany	Indoor	Quartz	1.1	Fluorescent tubes	298 ± 5 K	Evacuatable	Barnes et al. (1994)
RvG- ASIC University of East Anglia, Norwich, UK	Indoor	FEP and borosilicate glass	3.3	Fluorescent tubes	-55 to + 30 °C;	Atmosphere-ocean-sea-ice-snow simulation chamber	Thomas et al. (2021)
PACS-C3 PSI, Villigen, Switzerland	Indoor	FEP	27	Arc	Range 15–30 °C		Paulsen et al. (2005)
PACS-C3 PSI, Villigen, Switzerland	Indoor	FEP	9	Fluorescent tubes	Ambient	Mobile	Platt et al. (2013)
PACS-C3 PSI, Villigen, Switzerland	Indoor	FEP	9	Fluorescent tubes	Range 10–30 °C		Platt et al. (2013)
SAPHIR FZJ, Jülich, Germany	Outdoor	FEP	270	Sun	Ambient	Atmospheric chemistry at low concentrations, radical budgets	Rohrer et al. (2005a)

(continued)

Table 1.4 (continued)

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
SAPHIR-PLUS FZI, Jülich, Germany	Indoor	FEP	9	LED	0–50 °C	Plant chamber	Hohaus et al. (2016)
SAPHIR-STAR FZI, Jülich, Germany	Indoor	Quartz	2	Fluorescent tubes	Ambient	Continuous stirred reactor	
Large Aerosol Chamber FZI, Jülich, Germany	Indoor	Teflon	250	None	Ambient		Zhao et al. (2010)
<i>Other chambers worldwide</i>							
CSIRO, Sidney, Australia	Indoor	FEP	25	Fluorescent tubes	Ambient		Hynes et al. (2005)
Fudan University, Shanghai, China	Indoor	Teflon coated Stainless steel	4.5	No	Ambient		Li et al. (2015, 2017), Zhang et al. (2011a, b)
IAP, Beijing, China	Indoor	FEP	1	Fluorescent tubes	Ambient		Zhang et al. (2019)
CRAES, Beijing, China	Outdoor	FEP	56	Sun	Ambient		Li et al. (2021)

(continued)

Table 1.4 (continued)

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
Zhejiang University Smog chamber (CAPS-ZJU), Hangzhou, China	Indoor	FEP	3	Fluorescent tubes	Ambient	Dual	Li et al. (2020)
Guangdong University of Technology–dual-reactor chamber (GDUT-DRC), Guangzhou, China	Indoor	FEP	2	Fluorescent tubes	Ambient	Dual	Luo et al. (2020)
CAS, Guangzhou, China	Indoor	FEP	30	Fluorescent tubes	−10 to 40 °C		Wang et al. (2014)
RCEES, Beijing, China	Indoor	FEP	30	Fluorescent tubes	Controlled		Chen et al. (2019)
SHUSC, Shanghai University Smog Chamber, Shanghai, China	Indoor	FEP	1.2	Fluorescent tubes	Ambient		Qi et al. (2020)

(continued)

Table 1.4 (continued)

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
Tsinghua University, Beijing, China	Indoor	FEP	2	Fluorescent tubes	10–60 °C,		Wu et al. (2007)
Aarhus University, Aarhus, Denmark	Indoor	FEP	2	Fluorescent tubes	–15 to 26 °C		Kristensen et al. (2017)
Toyota, Aichi, Japan	Indoor	FEP	5	Fluorescent tubes	Ambient		Takekawa et al. (2003)
NIES, Tsukuba, Japan	Indoor	Teflon coated Stainless steel	6	Xenon arc lamps	0–40 °C	Evacuable and bakable up to 200 °C	Akimoto et al. (1979a, b, c)
NIES, Tsukuba, Japan	Indoor	FEP	0.7	Fluorescent tubes	–5 to 40 °C		Deng et al. (2021)
KIST, Korea Institute of Science and Technology, Seoul, Korea	Indoor	FEP	5.8	Fluorescent tubes	Ambient	Dual	Lee et al. (2009)
CLOUD, CERN, Geneva, Switzerland	Indoor	Stainless steel	26	LED	Controlled		Duplissy et al. (2010)
Carnegie Mellon, Pittsburgh, USA	Indoor	FEP	16	Fluorescent tubes	Controlled		Donahue et al. (2012)

(continued)

Table 1.4 (continued)

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
GeorgiaTech, Atlanta, USA	Indoor	FEP	12	Fluorescent tubes	Controlled	Dual	Boyd et al. (2015)
CalTech, Pasadena, USA	Outdoor	FEP	28	Fluorescent tubes	18–50 °C		Cocker et al. (2001)
CalTech, Pasadena, USA	Outdoor	FEP	65	Sun	Ambient		Leone et al. (1985)
UC Irvine, Irvine, USA	Indoor	Stainless steel	0.5	Hg and fluorescent tubes	Controlled	–10 to 70 °C	De Haan et al. (1999)
UC Irvine, Irvine, USA	Indoor	FEP	5		Controlled		Nguyen et al. (2011)
UC Riverside, Riverside, USA	Indoor	FEP	90	Ar-Xe arc lamp	5–45 °C		Carter et al. (2005)
UC Riverside, Riverside, USA	Indoor	FEP	6.6	Fluorescent tubes	Ambient		Lim and Ziemann (2005)
UC Riverside, Riverside, USA	Indoor	Teflon coated aluminium	5.6	Xenon arc	Ambient	Evacuatable	Beauchene et al. (1973), Winer et al. (1980)
UNC, Pitsboro, USA	Outdoor	FEP	135	Sun	Ambient	Half-cylinder-shaped dual chambers	Lee et al. (2004)

(continued)

Table 1.4 (continued)

Chamber name and location	Indoor/Outdoor	Wall material	Volume (m ³)	Light source	Temperature	Comment	References
Pacific Northwest National Laboratory, Richland, USA	Indoor	FEP	10.6	Fluorescent tubes	Controlled		Liu et al. (2012)
UNC, Chappel Hill, USA	Indoor	FEP	9	Fluorescent tubes	Ambient		Smith et al. (2019)
CU Boulder, Boulder, USA	Indoor	FEP	8	Fluorescent tubes	Ambient		Krechner et al. (2016, 2017)
Harvard/HEC, USA	Indoor	FEP	4.7	Fluorescent tubes	Ambient		King et al. (2009)
NCAR, Boulder, USA	Indoor	FEP	10	Fluorescent tubes	Ambient		Fry et al. (2014)
University of Florida UF-APHOR, USA	Outdoor	FEP	52	Sun	Ambient	Half-cylinder-shaped dual chambers	Im et al. (2014)

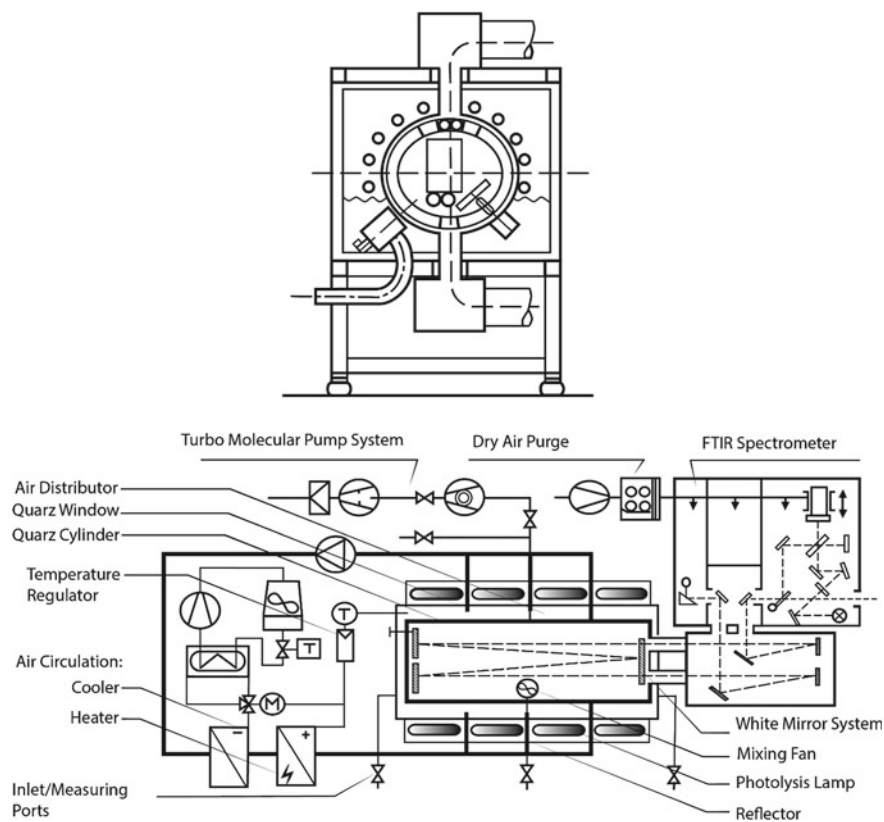


Fig. 1.4 Example of a 1 m³ indoor quartz chamber irradiated with UV fluorescent tube—the QUAREC chamber from the Bergische Universität Wuppertal–Germany. (© Bergische Universität Wuppertal)

1.2.2 Simulating Gas Phase Mechanism, Radical Cycles and Secondary Pollutant Formation

Studies on the formation of secondary pollutants are generally conducted in large outdoor chambers to avoid potential artefacts linked to a lack of realism in the irradiation and to minimize radical losses or conversion on the walls. Tropospheric ozone production studies were hence the first to benefit from chamber application. Nevertheless, for those studies to be of use for general modelling it is necessary to disentangle chamber effects from directly applicable results. Such an approach has led as early as the late 1970s to the first ozone isopleth diagrams, linking precursor levels to ozone production (Dodge 1977; Jeffries et al. 2013). Interestingly, because of the focus on photooxidants which is mostly driven by air quality legislation, operational model evaluation is often conducted by comparison with the results arising

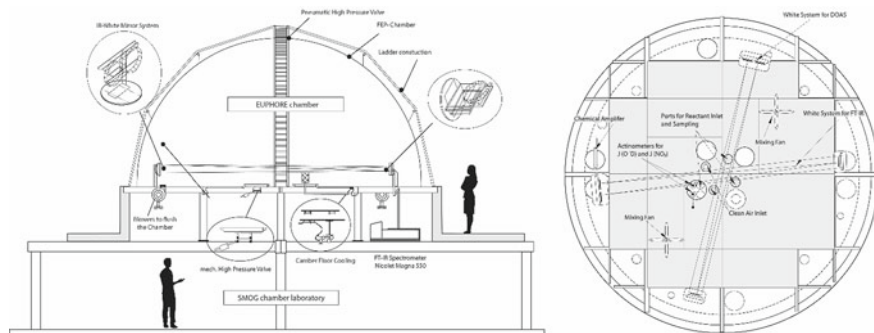


Fig. 1.5 Example of a large outdoor Teflon® chamber: the EUPHORE chamber–Valencia, Spain. (© EUPHORE)

from experiments conducted in these types of large chambers (Carter et al. 1979; Wagner et al. 2003; Bloss et al. 2005a, b; Carter 2008; Parikh et al. 2013).

Such chambers are made of FEP Teflon film, generally, several hundreds of cubic-meters in volume and are often installed on the roof of a dedicated laboratory (e.g. EUPHORE in Valencia, Spain Fig. 1.4 or Helios in Orleans, France) or in dedicated shelter structures (e.g. SAPHIR in Jülich, Germany Fig. 1.9 or UNC in North Carolina, USA). Because of their size and their outdoor installation, these facilities generally involve through-wall connections and inlets to connect the chamber with a measurement laboratory often located below. They also include devices such as a retractable roof to protect them from rain and wind. Temperature control cannot be achieved in such chambers and air inside the chamber may be heated by metal plates underneath the chamber when they are exposed to sunlight during the experiment. This effect is reduced if there is no direct contact of the metal plate with the chamber film and can be further reduced if the metal plate is cooled. Interestingly, even if their size is a significant advantage to minimize wall effects (on both gas phase and particulate phase), wind induced movements of the Teflon film lead to charge build-up that has the tendency to strongly reduce the physical lifetime of particle by drawing them to the wall (McMurry and Grosjean 1985) (Figs. 1.5 and 1.6).

1.2.3 Aerosol Processes

Originally considered as a technical problem during early smog simulation experiments, secondary organic aerosol (SOA) formation has since attracted very large interest from the scientific community. The availability of instruments such as Scanning Mobility Particle Sizers (SMPS), for the determination of particle number and size distribution with a time resolution of minutes, helped to promote the rapid development of experimental studies of SOA formation. This trend was further increased when mathematical formalisms were proposed to extrapolate the SOA yield from the

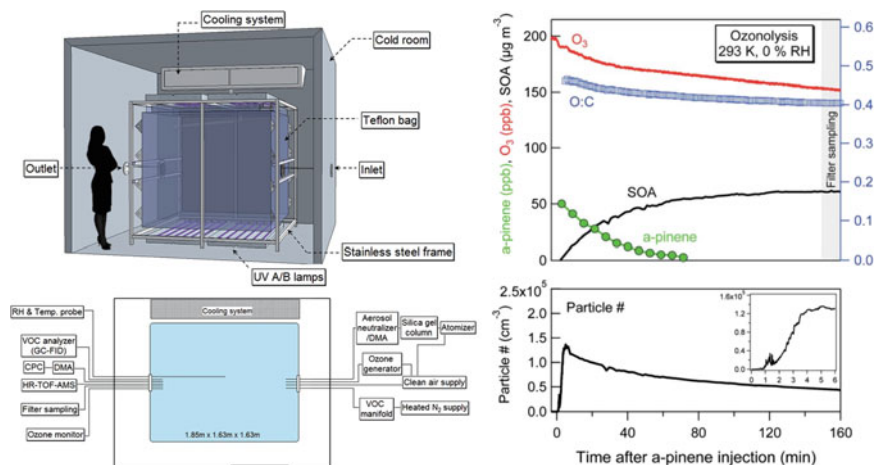


Fig. 1.6 Left: Example of a medium size indoor Teflon® chamber irradiated with UV fluorescent tubes/Right: Typical SOA production from terpene ozonolysis experiment (adapted from Kristensen et al. 2017)

high precursor concentrations used in chamber experiments to atmospheric conditions (Odum et al. 1996). The volatility basis set (VBS) formalism proposed by Donahue et al. (2006) was especially successful in providing a parameterization that could be inserted in models (3D included) and has triggered a renewed interest for chamber experiments from the modelling community. Both medium size and large chambers, as well as indoor and outdoor facilities, are regularly used for SOA experiments. Due to the multiphase nature of the processes studied and their even greater non-linearity, there is a general effort to reduce the starting concentration of the precursor to the ppb range (and sometimes below) in order to perform experiments at atmospherically relevant chemical conditions. These low concentrations make the results of these experiments very sensitive to wall effects on the gaseous species, such as wall loss of compounds that could normally participate in the aerosol mass or, on the other hand, the release of semi-volatile species. Further, physical wall losses of particles can also be significant. The quantitative characterization of these wall effects is still an open topic that requires a widely applicable formulation (see Chap. 2). It also depends highly on the properties of the wall (conductivity, permeability, reactivity, porosity...) in a context where the mechanisms involved are not yet well understood. Consequently, the combined use of several types of chambers, different in size but also made from different materials (Teflon film, glass, steel, aluminium...), is highly desirable for SOA experiments conducted at more realistic atmospheric concentrations of precursor gases. In parallel, a significant quantity of work has been conducted to better represent semi-volatile wall losses in this diversity of chambers (La et al. 2016; Krechmer et al. 2017; Lamkaddam 2017).

The contribution of simulation chambers to the understanding and quantification of SOA and related impacts is not limited to yield measurements. A wide body of

work has focused on both online and offline chemical characterization with the aim of understanding the chemical composition of the SOA fraction but also the chemical processes that govern the formation and aging of organic aerosol. As a result of the amount of work carried out in medium size chambers, important breakthroughs have been made in these topics such as the identification of oligomerization processes in the aerosol phase (Kalberer et al. 2006), the chemical trends followed by oxidation during SOA aging (Jimenez et al. 2009; Ng et al. 2011a, b; Kourtchev et al. 2016), or the importance of auto-oxidation processes for the formation of SOA precursors (Ehn et al. 2014).

New particle formation was long considered as a barely controllable step in the formation of SOA during simulation chamber experiments. For reproducibility purposes, in most of the studies focusing on aerosol yield, it is hence recommended to use seed aerosol as a condensation medium in order to avoid nucleation. Nevertheless, dedicated chambers—often exhibiting a very low level of electrostatic charges on the wall—have been used to investigate this important process that is possibly controlling the number of cloud condensation nuclei in some parts of the atmosphere (Bonn et al. 2002; Kiendler-Scharr et al. 2009a, b; Kirkby et al. 2011, Boulon et al. 2012). One of the challenges in studying the early steps of nucleation in simulation chambers is, on the one hand, the ability to measure clusters and particles in the range of 1 to 3 nm and, on the other hand, the reduced lifetime of particles smaller than 20 nm in enclosed vessels (see Sect. 2.5 for particle wall losses analysis). Indeed, simulation chambers easily allow for aerosol lifetimes of several hours to a few days for particles in the range of a few hundreds of nanometers but due to their very high diffusivity, particles in the range of a few nanometer exhibit lifetimes in the range of a few minutes only.

Because of the importance of nucleation related processes, a dedicated facility was set-up at CERN: the CLOUD (Cosmics Leaving Outdoor Droplets) experiment. The CLOUD chamber is a stainless steel atmospheric simulation chamber of 26.1 m³ (Duplissy et al. 2010; Voightländer et al. 2012) operating under drastically clean conditions and installed in the T11 beamline at the CERN Proton Synchrotron. In order to study the effect of cosmic rays on nucleation, the chamber can be exposed to a 3.5 GeV/c positively-charged pion (π^+) beam from a secondary target. The results from this atmospheric simulation chamber have led to significant advances in the understanding of nucleation including the elimination of the role of sulfuric acid alone as a nucleating agent, some insight on the effect of cosmic rays and the role of low volatility products from biogenic oxidation in initial cluster formation.

As aerosols refer to the particulate and gas phase, the investigation of aerosol processes in atmospheric simulation chambers also includes studies of heterogeneous processes. Prominent examples of systems studied include the chemical aging of aerosols and formation of brown carbon (e.g. Laskin et al. 2015) and the uptake of ozone on organic aerosol such as SOA formed from limonene ozonolysis (Leungsakul et al. 2005; Zhang et al. 2006). The N₂O₅ uptake coefficient on different particle types and the influence on gas phase oxidant levels were excessively studied in the Jülich indoor aerosol chamber (Mentel et al. 1996; Folkers et al. 2003; Anttila et al. 2006). More recently it was shown in atmospheric simulation chambers that levoglucosan,

traditionally utilized as a source tracer for biomass burning aerosol, is reactive in the atmosphere (Hennigan et al. 2010, 2011; Sang et al. 2016; Bertrand et al. 2018; Pratap et al. 2019).

1.2.4 *Cloud Processes*

While “cloud chambers” have existed for a very long time, mostly to study the microphysics of fog and clouds, the past few decades have seen emerging chamber facilities which can generate clouds and fog under sufficiently clean conditions that multiphase chemistry, transformation at the droplet interface and cloud microphysical processes can be studied (Stehle et al. for the DRI chamber 1981; Hoppel et al. for the CALSPAN chamber 1994; Möhler et al. 2001 for the AIDA chamber; Duplissy et al. 2010 for the CLOUD chamber; Wang et al. 2011 for the CESAM chamber; Chang et al. 2016 for the Pi Chamber). All of these chambers are made of metal—mostly stainless steel (except for AIDA where the walls are made of aluminium)—because one of the most common protocols to generate a cloud is to perform a quasi-adiabatic expansion through a relatively fast decrease of the total pressure (from a few second to a few minutes) with or without controlling the wall temperature. For instance, the AIDA chamber allows for generating liquid droplets, mixed-phase (droplet and ice) and pure ice clouds. Further details can be found in Sect. 8.1 (Fig. 1.7).

These facilities have opened the door for realistic studies of cloud microphysics in the laboratory. The studies, which have been enabled due to careful control of the initial and boundary conditions, include investigations into the cloud condensation nuclei (CCN) and ice nucleation activity of various aerosol particles (Wagner et al. 2011; Henning et al. 2012; Hoose and Möhler 2012), homogeneous freezing of supercooled solution droplets (Möhler et al. 2003), scattering properties of ice crystals (Järvinen et al. 2014; Schnaiter et al. 2016), and the effects of non-precipitating water clouds on aerosol size distributions (Hoppel et al. 1994).

In parallel, a whole field of activity has been opened with the ability to study chemical transformations at the interface of droplets or even in the suspended aqueous phase. Using this approach, sulfate formation from the multiphase oxidation of SO₂ has clearly attracted the most attention (Stehle et al. 1981; Miller et al. 1987; Lamb et al. 1987; Hoyle et al. 2016), but more recently, aqueous SOA formation from isoprene oxidation products (Brégonzio-Rozier et al. 2016) and brown carbon formation from fog processes of functionalized organics (De Haan et al. 2018) have also been investigated.

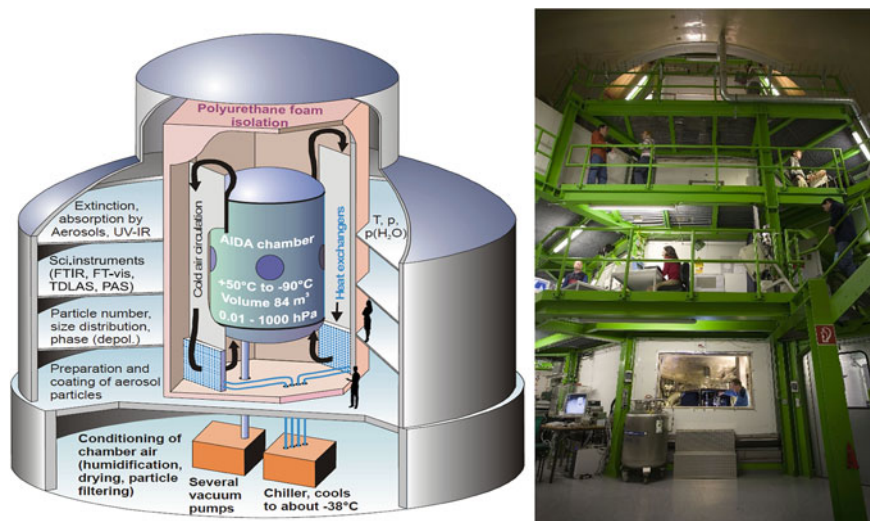


Fig. 1.7 Left: The AIDA facility at the Karlsruhe Institute of Technology with an 84 m³ aluminium chamber. Trace gas, aerosol and cloud experiments can be performed in a wide range of atmospheric temperatures (+60 °C to -90 °C), pressure (1–1000 hPa) and humidity (sub- and supersaturated with respect to liquid water and ice) conditions). Right: Typical evolution of pressure, temperature, relative humidity, and cloud droplet diameter for an adiabatic expansion experiment in AIDA

1.2.5 Characterization and Processing of Real-World Emissions

The development of atmospheric chemical mechanisms has been based on chamber studies of atmospheric oxidation of individual compounds. Hundreds of species have been studied following this approach and have contributed to the building of detailed chemical schemes, such as the Master Chemical Mechanism MCM (website: mcm.york.ac.uk). This effort is still ongoing to take into account new emissions and refine the chemical module of large-scale models. Nevertheless, in parallel, chamber studies that represent more realistic and more complex conditions are required to close the gap between well controlled but simplified laboratory experiments and observations in the real atmosphere.

Chamber studies, previously described here, have focused on chemical processes occurring in the gas and aerosol phases and have usually been limited to the simplified oxidation conditions and systems of selected precursors. More recent studies on real emissions from combustion sources such as engines and wood-burning stoves, or from natural emission sources such as plants or mineral dust, raise interesting possibilities for more relevant investigations of atmospheric processes.

In these studies, chambers are coupled to real emission sources (plant chambers, engines, wood burners, cooking stoves...) to study systems of real-world complexity. As much as one loses the ability to fully understand processes because of the

complexity of the starting mixtures, one gains in the realism of the impact and the enhanced comparison with field measurements.

Experiments using real-world emissions involve complex sources that are either so intense that they need to be diluted before being added to chambers (e.g. engines, wood burners, cooking stoves) or do not require dilution (e.g. plants, sea spray, air fresheners and other household products). Approaches to ensure the quantitative transfer of all compounds of such complex emission blends into atmospheric chambers are described in detail in Chap. 5.

Concerning the first category, these experiments involve primary pollution sources whose aging is studied because of a potential formation of secondary pollution worsening their primary effect. The experimental challenges here are to

- a. reproduce the atmospheric dilution of primary emission (both gaseous and particulate matter) while remaining in measurable concentrations: generally, a dilution factor ranging between 100 and 1000 are used (Platt et al. 2013, 2017; Gentner et al. 2017; Pereira et al. 2018)
- b. establish a chemical system mimicking atmospheric aging over a few days.

Large and medium size chambers can be used for these studies. For example, Geiger et al. (2002) have connected a diesel engine fuelled with various diesel fuel formulations and mounted on a motor test bed directly to the EUPHORE chamber. In the dual outdoor simulation chambers, VOC mixtures containing a fixed ratio of *n*-butane, ethene and toluene were irradiated by natural sunlight in the presence and the absence of diesel exhaust. In this case, the large volume of the EUPHORE chamber (ca. 200 m³) removed the need for a dilution system. For smaller simulation chambers (Chirico et al. 2010; Pereira et al. 2018; Platt et al. 2013) a conservative dilution system is needed to reduce the concentrations while keeping constant the various ratios between gaseous and particulate species, volatile and semi-volatile species. To do so, a specific aerosol diluter and heated lines are used. To preserve the efficiency of the atmospheric processes, prescribed VOC-to-NO_x ratios are used which often require the addition of a VOC such as ethene, which is chosen for its ability not to add to the particulate mass during its oxidation. Aging is, for example, evaluated using the OH exposure index, defined as the cumulative OH concentration over the course of the experiment. The calculation of OH exposure requires the use of an OH tracer such as deuterated butanol-d₉ (Barnet et al. 2011) or the direct measurement of OH (e.g. Zhao et al. 2018) (Fig. 1.8).

These studies have demonstrated that, when considering car emission related fine particles, secondary pollution was as important as primary pollution and sometimes larger (Geiger et al. 2003; Bahreini et al. 2012; Platt et al. 2013, 2017; Gentner et al. 2017). In particular, the content of intermediate volatility organic compounds (IVOC) has been identified as critical in the ability to produce SOA (Pereira et al. 2018). The work in simulation chambers has allowed testing of the various types of vehicles, engines or fuel formulations that were already available on the market but, the interest that this methodology has raised among car manufacturers, allows one to hope for testing of future technology before its widespread deployment in vehicles.

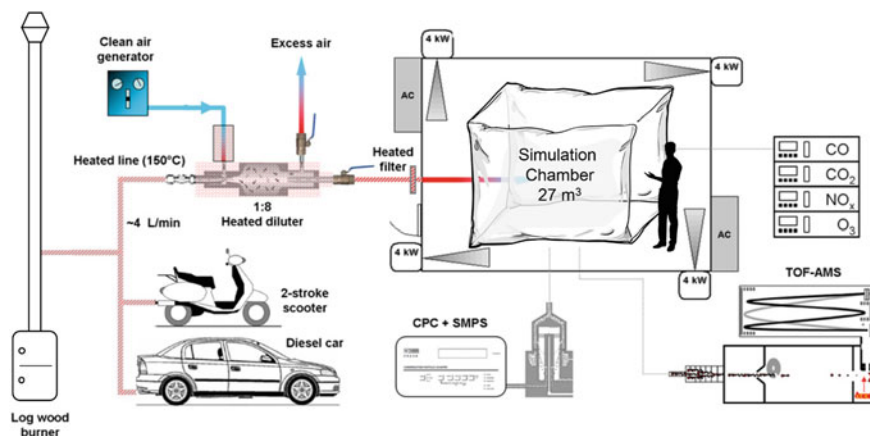


Fig. 1.8 Use of the PSI 27 m³ Teflon chamber for investigating various real-world emissions transformation in the atmosphere. (Figure reused with permission from Heringa et al. (2012) Open access under a CC BY 3.0 license, <https://creativecommons.org/licenses/by/3.0/>)

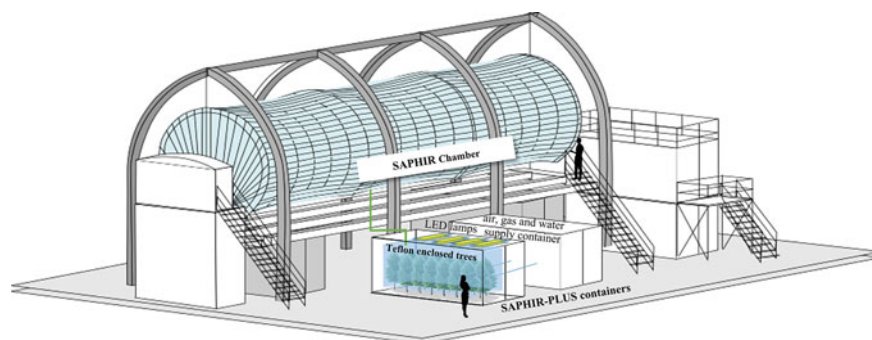


Fig. 1.9 SAPHIR-PLUS the combination of one of the largest outdoor simulation chamber (SAPHIR–Forschungszentrum Jülich, volume: ca. 270 m³) with a controlled plant growing unit

A similar methodology can be applied to biomass combustion emissions. Considering the importance of this family of emissions, sources such as in-house open fires, agricultural burning, modern stoves or even barbecue emissions have been injected in a simulation chamber and aged in order to better quantify the extent of secondary pollution relatively to primary emission (Tiitta et al. 2016; Bertrand et al. 2017; Bhattu et al. 2019). Not only do these studies allow evaluation of the environmental impacts of combustion of various fuels (e.g. logwood, pellet, straw), types of combustion technology (e.g. stoves) and the various burning regimes (such as flaming or smouldering), but they also allow identification of molecular tracers and mass spectral signatures that can be monitored in the field to improve emissions inventories.

For experiments involving the atmospheric processing of plant emissions, the key challenge is not the dilution as these emissions are diffuse enough, but rather the preservation of their representativeness. Indeed, as living organisms, plants are sensitive to their environmental condition and any unwanted factors such as water stress, mechanical stress, biotic stress, oxidative stress or other abiotic stress from air composition may affect the composition and amount of their emissions (e.g. Kleist et al. 2012; Mentel et al. 2013; Wu et al. 2015; Yli-Pirilä et al. 2016; Zhao et al. 2017). Consequently, for studies involving plants, the plant growing facility as well as the emission transfer system have to be the subject of extreme care.

In SAPHIR-PLUS for example (see Fig. 1.9, Hohaus et al. 2016), the photo-oxidation of *Pinus sylvestris* L. (Scots pine) emissions were reacted and aged by ozonolysis in the presence of sunlight (Gkatzelis et al. 2018) which has allowed parameterization of the SOA production from these real plant emissions following the volatility basis set (VBS) formalism (Donahue et al. 2006). In a 9 m³ temperature controlled Teflon simulation chamber, run in batch mode at the University of Eastern Finland, Failo et al. (2019) studied SOA formation from healthy Scots pine emissions and from the same plants infected with aphids. The aphid stressed pine were shown to emit more linear sesquiterpenes than healthy ones with significant effects on the SOA yields. Wyche et al. (2014) investigated in the Manchester Aerosol Chamber (MAC), the differences in SOA formed from predominantly terpene versus predominantly isoprene emitters. So far only very few studies have examined SOA production from the full range of VOCs made by plants. Since it was shown that the individual contributions of VOC in mixtures interact in non-linear ways in SOA formation mechanisms (Kiendler-Scharr et al. 2009a, b; McFiggans et al. 2019), there is a strong need for more studies exploring plant emissions.

1.2.6 Mineral Dust

aerosols are another key player in the atmospheric system. These particles contribute to the aerosol radiative effect and can act as cloud condensation nuclei (CCN) as well as ice nucleating particles (INPs). Mineral dust particles can deliver soluble elements needed for the development of oceanic life and eventually modify the CO₂ content of the atmosphere. Altogether, these kinds of aerosol particles affect Earth's weather and climate. Desert dust also affects human health, as an irritating agent at high concentrations causing respiratory diseases, as well as a vector for bacteria, viruses and possibly for severe infections like meningitis.

During transport, mineral dust can mix with air pollution and undergo chemical transformations that may affect their basic properties (composition, optical properties, CCN/IN activities, solubility...) and therefore their atmospheric impacts. Further, the multiphase chemistry occurring at their surface may also affect air composition. All these reasons have recently led a small number of research groups in the chamber community to apply the experimental simulation methodology to this science topic. This application implies solving various issues.

The first issue is the representativeness of the generated dust aerosol with respect to the atmosphere. Airborne mineral dust is a mixture of several minerals whose proportions change depending on the properties of the parent soil and wind speed. It forms an aerosol of an extended size distribution (extending from hundreds of nanometers to tenths of micrometers) that does not necessarily reflect the mineralogy of the soil due to the size-dependence fractionation between the soil and the aerosol phases that occurs at emission. There is hence a technological challenge in reproducing the dust generation from the soil process so that both the mineralogical composition and the size distribution are realistic (see Sect. 5.2). The global diversity of the mineralogical composition of natural parent soil is not reproduced by the commercially available minerals or standard mixtures. As much as possible, research tries to face this diversity by generating dust from natural soil collected across the world (Linke et al. 2006; Möhler et al. 2008a; Connolly et al. 2009; Wagner et al. 2012; Di Biagio et al. 2014, 2017a, b, 2019; Caponi et al. 2017), complementing and augmenting the many studies with model mineral dust such as Arizona Test Dust (Möhler et al. 2006, 2008a, b; Connolly et al. 2009; Vlasenko et al. 2006) or pure minerals such as illite (Möhler et al. 2008a, b) kaolinite (Tobo et al. 2012), hematite (Hiranuma et al. 2014) or Feldspar (Mogili et al. 2006, 2007; Atkinson et al. 2013).

Another critical issue for the study of mineral dust in simulation chambers is the reduced lifetime of these aerosols. Indeed, simulation chambers easily allow for aerosol lifetimes of several hours to a few days for particles in the range of a few hundreds of nanometers, but particles in the range of several micrometers undergo rapid sedimentation. As a consequence, in the absence of active resuspension processes, their lifetime in enclosed vessels is reduced to a few minutes only. This makes it difficult to study chemistry at the surface of the coarse fraction of mineral dust, but it is an advantage when one tries to reproduce the physical aging of dust plumes in the atmosphere. In fact, chamber experiments of a couple of hours duration can reproduce modifications to the size distribution of airborne dust that takes place over 2–3 days of transport (Di Biagio et al. 2017a, b). Chambers are therefore an emerging tool of choice to study the hygroscopicity and optical properties of mineral dust or the chemistry in the presence of the fine fraction only.

To date, most of the published results from chamber studies involving mineral dust have focused on their direct and indirect radiative effect. A large number of ice nucleation studies have been carried out at the AIDA chamber and LACIS (Leipzig Aerosol Cloud Interaction Simulator) on surrogate dust left bare (Möhler et al. 2006, 2008a, b; Tobo et al. 2012; Hiranuma et al. 2014; DeMott et al. 2015; Niedermeier et al. 2011, 2015; Hartmann et al. 2016) or covered with inorganic (Augustin-Bauditz et al. 2014; Niedermeier et al. 2011; Wex et al. 2014) and organic layers (Möhler et al. 2008a, b). In the CESAM chamber, most of the research to date has focused on optical properties and the derivation of complex refractive indexes in the long wave spectral ranges (Di Biagio et al. 2014, 2017a, b) and in the UV–visible (Di Biagio et al. 2019, Caponi et al. 2017).

To date, the number of studies of chemical reactivity at the surface of mineral dust in simulation chambers is rather limited due to the above-mentioned difficulties. They mostly involved ozone loss on the particles (Mogili et al. 2006) or SO₂ uptake and reactivity (Zhou et al. 2014).

1.3 Bridging the Gap Between Laboratory and Field Studies

Simulation chambers have been also used for the benefit of field experiments and long-term atmospheric monitoring (Kourtchev et al. 2016). These cross-community activities have first concerned instrumental development with a number of high technology new techniques being developed or tested at simulation chambers (see also 1.5). Prominent among these types of studies is the development of new techniques dedicated to atmospheric radical measurement (Schlosser et al. 2007; Onel et al. 2017), new techniques involving advanced optical setups such as optical cavities (Varma et al. 2009, 2013), the development of new advanced mass spectrometry instruments (Docherty et al. 2013) and chromatographic procedures for the elucidation of the aerosol organic fraction (Rossignol et al. 2012a, b).

1.3.1 Tracers and Sources of Fingerprint Studies

The use of simulation chambers for the benefit of field studies also includes the identification of specific signatures for emission sources (especially for aerosol mass spectrometry—see Aiken et al. 2008; Mohr et al. 2009; Kiendler-Scharr et al. 2009a, b; Zhang et al. 2011a, b; Schwartz et al. 2010). It also involves the identification of molecular tracers characteristic of specific processes. In this case, the ability of chambers to study specific processes is valuably used to separate the effect of the various potential oxidants or conditions. When well characterized, and found to be sufficiently unreactive in the atmosphere, these tracers are then searched for in the field to apply advanced apportionment procedures with the aims of not only elucidating the extent of primary sources but also of secondary processes (Jaoui et al. 2007; Kleindienst et al. 2007, 2012; Zhang et al. 2012).

In addition, important work has been carried out in characterizing the atmospheric tracers of primary sources such as levoglucosan or guaiacol (Hennigan et al. 2010; Bertrand et al. 2018; Pratap et al. 2019) that were initially thought fairly unreactive. This includes the use of stable isotopes as tracers for the extent of chemical processing (Sang et al. 2016; Gensch et al. 2014).

1.3.2 Instrument Comparison Campaigns

In addition to activities which involve generally one or only a few groups, large instrument comparison campaigns gather the wider atmospheric science community around chambers to characterize both established and emerging techniques using the ability of simulation chambers to precisely control the environmental conditions, while allowing different instruments to simultaneously sample from the same air mass. Suspected artefacts can hence be intentionally amplified and the sensitivity of the related techniques can be investigated and quantified. High precision water vapor measurement (Fahey et al. 2014), NO_x and NO_y measurements (Fuchs et al. 2010), oxygenated species measurements (Wisthaler et al. 2008; Apel et al. 2008; Thalman et al. 2015; Munoz et al. 2019), radical measurements (Schlosser et al. 2007; Fuchs et al. 2010; Fuchs et al. 2012a, b; Ródenas et al. 2013; Onel et al. 2017) or radical reactivity measurements (Fuchs et al. 2017) have been compared in large campaigns at chambers during the last 15 years.

1.3.3 Field Deployable Chamber

Recently a very innovative approach which combines the use of a simulation chamber with field studies has been developed both in Patras (Greece) and in Carnegie Mellon Institute (USA). It involves the use of portable simulation chambers directly in the field. This strategy is based upon a concept experiment: use ambient air as a starting point and allow the study of the evolution of atmospheric particulate matter at timescales longer than those achieved by traditional laboratory experiments (Kaltsonoudis et al. 2019).

This type of study can take place under more realistic environmental conditions but they could appear as being contrary to the whole simulation chamber experiment concept i.e. simplify and control the chemical system to better understand it. To solve this apparent contradiction, the group that is developing this new approach has developed a dual chamber strategy: after careful characterization of both chambers and so after verifying that they are producing comparable results, both are filled with the ambient being studied but one is “perturbed”. The perturbation can consist of an additional oxidant injection such as ozone, addition of OH sources such as HONO or H₂O₂, or the addition of a compound potentially modifying the aerosol formation scheme such as α -pinene (Kaltsonoudis et al. 2019). The information on the chemical state of the sampled air is then deduced from the differential analysis of the results from the perturbed and control chambers (Fig. 1.10).

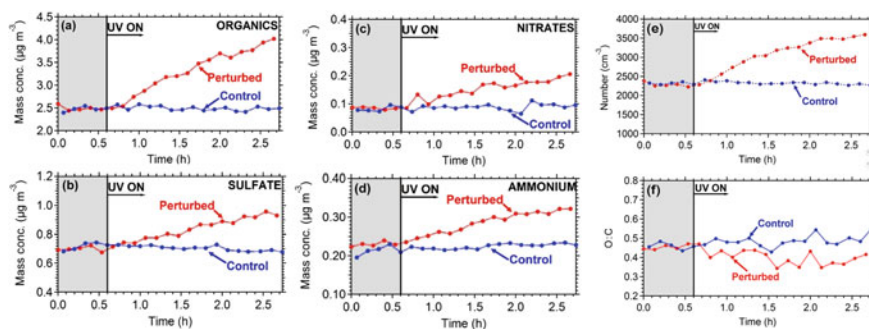


Fig. 1.10 Results from the operation of a dual field deployable simulation chamber during a campaign in Pittsburg (USA). One chamber is perturbed with the addition of HONO as an additional OH source **a** submicronic aerosol mass **b** Sulfate content of sampled aerosol in both chambers as measured by an AMS **c** Nitrate aerosol content **d** Ammonium aerosol content **e** submicronic particle number concentration **f** Oxygen-to-carbon ratio in the organic fraction of the aerosol as measured by an AMS. (Reused with permission from Kaltsonoudis et al. 2019, open access under a CC BY 4.0 license, <https://creativecommons.org/licenses/by/4.0/>)

1.4 Emerging Applications

1.4.1 Air-Sea/Ice Sheet Interaction

Recently, even more specific installations have been developed across the simulation chamber community: a chambers dedicated to the elucidation of processes occurring at the air-sea interface. It consists of chambers that include a reservoir at their bottom where artificial or real sea water is kept under controlled conditions and in exchange with the atmosphere above. In Lyon (France) such a chamber has been developed and used to study the processes occurring in an organic film deposited at the water surface and potentially affecting the simulated atmosphere composition. From a modelled sea water containing, humic acid ($1-10 \text{ mg L}^{-1}$) as a proxy for dissolved organic matter, and nonanoic acid ($0.1-10 \text{ mM}$), a fatty acid proxy which formed an organic film at the air-water interface, this work has shown that a photosensitized production of marine secondary organic aerosol could occur (Bernard et al. 2016). These new results suggest that in addition to biogenic emissions, abiotic processes could be of importance for the marine boundary layer. In East Anglia (UK), the *Roland von Glasgow* Air-Sea-Ice Chamber (RvG-ASIC), named in honour of its late founder, allows users to simulate sea ice growth and decay in a controlled environment. The tank can be filled with artificial or natural seawater and can be capped with a Teflon sheet to reproduce an experimental atmosphere. Here the main challenge is to produce a realistic sea-ice from the cooling of the seawater tank (the whole facility can be temperature controlled from $+30$ to $-55 \text{ }^\circ\text{C}$). This new facility has allowed investigating the mechanisms governing the fate of persistent organic contaminants in sea ice. It has shown that sea ice formation results in the entrainment of chemicals

from seawater, and concentration profiles in bulk ice generally showed the highest levels in both the upper (ice–atmosphere interface) and lower (ice–ocean interface) ice layers making them available from transit toward other compartments or interface reactivity.

1.4.2 Health Impacts

Even though the need to understand atmospheric chemistry has always been significantly motivated by public health issues and solving these issues has been part of the rationale for building many simulation chambers, until very recently, studies directly focused on health were rather scarce. In early investigations, the carcinogenicity and mutagenicity of chamber products were mostly evaluated after sampling of the contents and applying rather targeted offline in-vitro tests such as the Salmonella typhimurium plate-incorporation test (Claxton and Barnes 1981; Pitts 1983). In the past ten years, important progress has been made with the rise of surrogate indicators to qualify and quantify the potential health impact of particles such as the Reactive Oxygen Species content (ROS) (Fuller et al. 2014). The development of the corresponding instrumentation (Campbell et al. 2019) operating at high time resolution (on-line) now opens the way to building links between these indicators and the detailed chemical analysis often performed in the chamber. The goal is a better chemical characterization of the actual molecules or molecular functions involved in the oxidative stress.

In parallel, many groups have connected their simulation chambers with online samplers to expose living organisms such as lung cells or epithelial cells to the secondary pollutants produced in chambers (Savi et al. 2008; Mertes et al. 2013) in an attempt to understand the mechanisms that link cell toxicity with smog chemical and physical composition. This approach has led to important advances, especially when coupled with chamber experiments involving real world emissions (Künzi et al. 2013, 2015; Nordin et al. 2015). New directions have been explored by a few groups (Coll et al. 2018) which involve the use of simulation chambers for the long-term exposure (several days to several weeks) of living organisms such as murine models while complying with ethical standards. This new development requires overcoming substantial technical issues such as the stable and controlled production of secondary pollution over several days in a chamber. Their methodological research is pointing toward the use of indoor simulation chambers operated in batch mode. Development of such platforms in full cooperation with colleagues in the toxicology and medical communities may bring this health-related research to a better integration of the living body's functioning in the understanding of its response to air pollution.

1.4.3 *Bioaerosols*

Bioaerosols have been studied for over a decade in cloud chambers to investigate their potential ice nuclei activity (Möhler et al. 2008b). Given the public health problems associated with bioaerosol contamination and the many unknowns about the survival and transformation of bioaerosols, such as bacteria, in the atmospheric environment, innovative chamber work has recently started to address these issues (Amato et al. 2015; Brotto et al. 2015). These studies have led to the development of an indoor simulation chamber at the University of Genoa (Italy) where viable bioaerosol can be directly collected using Petri dishes without perturbing the course of the experiments while, in parallel, being online monitored by more classical techniques such as WIBS (Massabò et al. 2018). The goal is to derive parameterization of survival and activity of bioaerosols to eventually model the geographical extent of their contamination area.

1.4.4 *Cultural Heritage*

Works of art, with highly sensitive colours and materials, may be exposed to harmful levels of particulate matter in both indoor and ambient (i.e. outdoor) environments. Over time, these particles can deposit onto the surface of the artwork, which may influence the perceived colour. Reports over the concern of colour degradation to paintings, buildings, and other pieces of cultural heritage due to exposure to air pollution, acid rain, and other environmental factors have existed since at least the late 1800s due to London smog events (Brommelle 1964). However, the physical processes that connect exposure to particulate matter and the corresponding change in perceived colour are unknown, and first attempts to experimentally quantify the impact of particulate matter on painted works of art are only now emerging. The FORTH art exposure facility makes such an approach by developing protocols for the exposure of artwork to known levels of air pollutants and quantifying the effects of exposure using a portable colourimeter model WR-10 (FRU). Further developments in this emerging field will benefit from combining the expertise of exposure chamber approaches and atmospheric simulation chambers.

1.5 Considerations on the Design of an Atmospheric Simulation Chamber

The main objective of the guide is to serve as a reference for both new and current users of atmospheric simulation chambers. However, some readers may be considering the construction of a new chamber and this section is aimed at them. Additionally, it will provide to the new user, some insights into the design rationale of the chambers they will be working with.

This section mainly deals with the scientific issues and objectives that drive a particular chamber construction, but of course, practical limitations such as space, personnel and money will also influence chamber design. A particular focus is put on the requirements for the design of chambers dedicated to the exploration of atmospheric chemistry processes.

Atmospheric simulation chambers have several uses; firstly, they may be used to provide a controlled and realistic environment to simulate aspects of the real atmosphere or to test and compare field instrumentation. Secondly, chambers can be used as extended laboratory apparatus. For example, several hundreds of elementary reactions are involved in the complete oxidation of complex volatile organic compounds (VOC) such as isoprene (C_5H_8) or aromatic hydrocarbons. Some of these processes, particularly those occurring in the initial stages, can be studied individually by techniques such as laser flash photolysis or discharge flow, but many cannot. Atmospheric simulation chambers equipped with a wider range of instrumentation may either be able to directly measure rate coefficients, provide information on the yields of stable first-generation products, test entire chemical mechanisms or investigate aerosol chemistry. The main purpose of the experiments also strongly influences the design of the chamber.

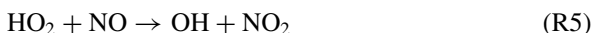
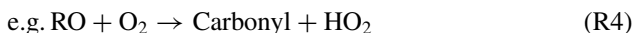
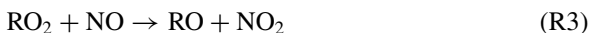
1.5.1 Chemical Regime of Simulation Experiments

Whatever the objective of the chamber, the primary applications are to processes in the Earth's *troposphere* (extending from surface to the tropopause, where tropopause height varies with latitude from ~10 km in polar regions to ~18 km in the tropics). In the troposphere temperatures range from ~220–320 K and pressures of ~100–1000 mbar are found. In addition, we are often interested in the interactions of emissions (biogenic or anthropogenic) with the atmosphere and the interactions of atmospheric pollutants with humans, animals, plants and the ocean. Most of these interactions take place within the *boundary layer*, typically the first kilometre or so of the troposphere and therefore for many applications, operation at pressures close to 1000 mbar is appropriate. However, there is obviously still a wide range of temperature variation within the boundary layer and so temperature variation may be an important goal in chamber design. Relative humidity also varies over a wide range in the troposphere and affects many physical and chemical processes in the atmosphere. Therefore, depending on the application of the chamber, precise control of humidity is also vital.

Besides variations in physical parameters, there are also significant variations in the chemical composition desired in the simulation experiments that will influence the chamber design. Most studies focus on regions of the atmosphere with significant VOC emissions. The chemical oxidation of VOCs often includes the same initial reaction steps; the reaction of a radical species, X, (where $X = OH, NO_3, Cl$ etc.) leads via abstraction or addition of the oxidant to an organic radical, R, which then rapidly adds O_2 to lead to an organic peroxy radical RO_2 .

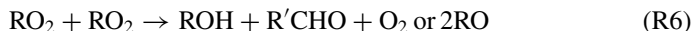


The atmospheric fate of the organic peroxy radicals depends on the relative abundance of concentrations of reaction partners such as nitric oxide ([NO]) and other peroxy radicals ([RO₂/HO₂]). In regions with high NO_x concentrations, the loss of RO₂ is typically dominated by the reaction with NO, generating an alkoxy radical (RO). The exact fate of the RO depends on its structure, but most often products are a carbonyl compound and hydroperoxyl radicals (HO₂). Further reaction of HO₂ with NO regenerates OH completing a reaction cycle (Fig. 1.10)



The by-product of the NO to NO₂ conversion in reactions (R3) and (R5) is ozone, a significant secondary pollutant. This radical reaction chain is the only relevant chemical source for ozone in the troposphere.

However, in environments with low NO_x concentrations (typically [NO] < 50 pptv) such as the marine boundary layer or remote tropical or boreal forests, radical recombination reactions become the dominant RO₂ loss channel.

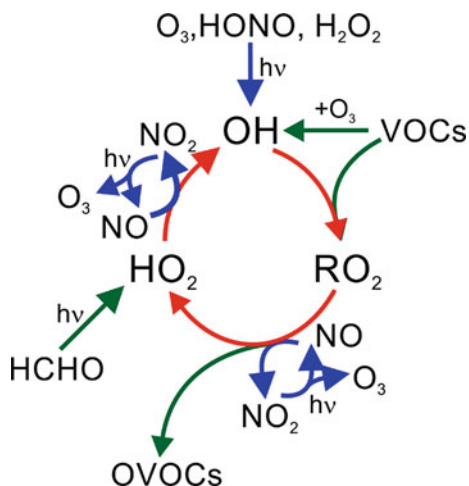


These reactions terminate the radical chain. For specific RO₂ radicals, isomerization reactions can be competitive. Products can be again RO₂ radicals that may decompose and thereby form other radical species such as HO₂ or highly oxygenated molecules could be eventually formed. For example, significantly enhanced OH concentrations are observed in high isoprene and low NO_x environments that can be explained by radical production from isomerization reactions of isoprene derived RO₂ (Peeters et al. 2014; Novelli et al. 2020).

Due to the importance of the fate of RO₂ radicals for the chemical reaction system that should be investigated in the simulation experiments, considerations about the NO_x concentration that can be achieved in the chamber is important and can have implications on the chamber design (Fig. 1.11).

The chemical composition of the troposphere is also impacted by surface interactions such as bulk and aerosol surfaces. The interaction with bulk solid surfaces

Fig. 1.11 Scheme of the radical reactions involved in atmospheric photochemical VOC oxidation and ozone production



can be easily replicated in many chambers. Some chambers (e.g. ISAC) are specifically designed to investigate interactions with liquid surfaces and sea-ice like the Roland Van Glasow Air-Sea-Ice Chamber at the University of East Anglia. Aerosols, primary or secondary, organic or inorganic, are the other main surfaces in the troposphere and studies involving aerosols and gas/aerosol/cloud interactions may require specific design criteria and instrumentation.

1.5.2 Chamber Size

Whilst there may be specialized chambers for the investigation of interactions with bulk surfaces, often bulk surfaces and their associated heterogeneous chemistry are minimized to avoid that experiments are impacted by chamber wall effects. Minimizing the surface to volume ratio (S/V) helps and might be the only way to suppress chamber wall effects, if experiments are performed at atmospheric concentrations of trace gases. For example, the large chambers EUPHORE (200 m^3) and SAPHIR (270 m^3) have spherical and cylindrical shapes, respectively, to minimize the surface to volume ratio and are advantageous compared to cuboid structures. Cuboid shapes are commonly used for Teflon chambers as they can be easily mounted, illuminated and physically accessed.

Most chambers have capabilities to inject reagents and maintain a homogeneous mixture by operating fans. Clearly, the specifications of fans need to match the chamber size to ensure efficient operation. The practical issues concerning logistics are beyond the scope of this chapter, but it is worth highlighting that large chambers such as AIDA, EUPHORE and SAPHIR have significant numbers of dedicated personnel and additional infrastructure facilities for example for clean air generation and power requirements.

As well as providing a more realistic environment for simulations, large chambers are ideal tools for field instrument comparisons. The volumes of gas sampled by some instruments make comparisons in small chambers impossible and generally there is more space for instruments. In situ comparisons in the real atmosphere have their advantages, but instrument comparisons in large chambers ensure, that all instruments sample the same chemical composition in a controlled environment and conditions can be systematically varied (e.g. Dorn et al. 2013; Fuchs et al. 2010, 2017; Fuchs et al. 2012a, b).

Whilst a small surface to volume ratio helps in ensuring that the chemical processes studied are indeed dominated by gas phase chemistry and ensures the best representation of atmospheric processes, this may not be required for other purposes of environmental chambers. For mechanistic or relative rate reaction kinetic studies, the rapid turnaround time of smaller chambers, where several experiments can be run per day, is far more efficient than performing such experiments in large chambers where studies may only be possible for good weather conditions in the case of outdoor chambers and may be limited to one experiment per day. Smaller chambers (particularly if made from glass or metal) can be rapidly evacuated (and in some cases heated) to clean the surfaces or can be even physically cleaned. Surfaces can be coated to minimize wall effects. Furthermore, many small chambers are operated in steady state conditions contrary to the batch mode operation of large chambers.

1.5.3 Materials

In general, there are three types of materials used in chambers: Teflon (or equivalent), borosilicate glass, quartz, or stainless steel (see Table 1.4). All materials have their advantages and disadvantages with respect to surface properties and physical parameters (e.g. T , p) that can be regulated in the chamber. Depending on the purpose of the chamber, the possibility to simulate e.g. pseudo-adiabatic cloud expansion, ultra-clean air conditions, or photolytic conditions representative of the troposphere is a key driver of choices of material used.

Teflon (or equivalent). Due to their large size, all large ($> \sim 80 \text{ m}^3$) chambers are constructed from fluoro-polymer plastics mounted on a metal frame. Such structures are light but fragile and need to be protected. Outdoor chambers like SAPHIR and EUPHORE have retractable protection, protecting the film from bad weather conditions, but also allow for experiments in the dark. The Helios chamber ($\sim 90 \text{ m}^3$) at CNRS-Orleans can be rapidly moved in and out of a permanent shelter. All of these chambers have a solid metal floor that can be used to place equipment such as FTIR mirrors and fans. In EUPHORE this forms part of the chamber surface and is cooled to prevent significant heating from solar radiation. In SAPHIR it is covered with Teflon and can be lowered for experiments such that the Teflon film does not have contact with the metal to avoid radiative transfer heating.

Teflon is also used in the construction of smaller chambers where glass or metal would be alternatives. Teflon has significant advantages in terms of cost. Additionally, as it is transparent, it is easy to fully illuminate the entire chamber with either solar or artificial light. Although Teflon is chemically inert, it is commonly observed that compounds can adhere to the wall and released in later experiments even if the chamber had been cleaned in between. For example, nitrous acid (HONO) is released, if humidified air is illuminated in Teflon chambers. The photolysis of HONO serves as a source of OH radicals, but also leads to an increase of nitrogen oxide species over the course of an experiment (Rohrer et al. 2005). The radical production from the chamber HONO source can be sufficiently high for performing OH oxidation experiments in large chambers as EUPHORE and SAPHIR (Fuchs et al. 2013). Smaller chambers can be manually cleaned, but this is not possible for larger chambers. As non-rigid structures, Teflon type chambers cannot be evacuated and are limited to operation at ambient pressures. Rather than evacuation, residual trace gases are removed by flowing clean gas through the chamber. For large chambers, this is typically done overnight. Smaller chambers can be enclosed in air-conditioned rooms to provide some degree of temperature control and variation.

Pyrex/Quartz Pyrex or quartz chambers are used for volumes of $\sim 1 \text{ m}^3$ or less. Within EUROCHAMP, the chambers at Wuppertal and Iasi are of cylindrical shape ($\sim 0.5 \text{ m}$ diameter) and have a volume of approximately 1 m^3 . The end flanges of both chambers are metal allowing for easy access to instrumentation and provide a fixed framework for mounting FTIR mirrors (similar structures are also used in some Teflon type chambers too). Due to the fragility of glass, the chambers are mounted on a vibration resistant framework. The advantage of quartz is that it allows for the transmission of shorter wavelength UV radiation compared to Teflon (e.g. radiation from mercury lamps emitting at 254 nm) which can be useful for specific radical generation methods.

Whilst pyrex/quartz chambers are limited in size, their small size allows to uniformly distribute artificial light sources around the chamber. The rigid construction also allows to evacuate the chambers, so that the chamber can be cleaned within a short time between experiments and it can be operated at sub-ambient pressure. Smaller chambers such as those at the National Centre for Atmospheric Research (NCAR) in Boulder, US, are surrounded with air-conditioned liquid baths to perform studies in which the temperature is varied. Quartz and pyrex are well characterized and reasonably inert surfaces. Evacuation (in combination with heating if available), provides rapid and efficient cleaning, in extremis, the end flanges of large chambers can be removed to allow for physical cleaning.

Metal Chambers are typically of cylindrical shapes and have volumes of the order of $1\text{--}6 \text{ m}^3$, with the exception of the 84 m^3 large AIDA chamber at Karlsruhe Institute of Technology. Metal chambers are typically constructed from stainless steel and have significant advantages in their robustness compared to other materials allowing for rapid evacuation/operation at reduced pressure. Several systems are also equipped with a temperature control system. Temperature control can be useful for two main purposes; firstly, simulating the temperature variation both within the boundary

layer at various latitudes/seasons and across the vertical extend of the troposphere; Secondly, elucidating the temperature dependence of chemical mechanisms.

For metal chambers, flanges with inlets for instruments are easy to install either in the main end flanges or elsewhere at the chamber. Although the end flanges of chambers can be large, they typically bow slightly if the chamber is operated at reduced pressure and therefore thought needs to be given on how to mount equipment requiring high spatial precision (e.g. multi-pass mirror) onto the end flanges.

The two significant disadvantages of metal as the construction material (besides the high S/V associated with the relatively small volume of most chambers) is the potential reactivity of the surface and the difficulty in generating a uniform light field. Surface effects can be accounted for (see Sects. 2.4 and 2.5) and efficient evacuation combined with overnight heating and/or oxidant exposure (e.g. O₃) ensures that the surface remains uniform over the course of an experimental campaign (see Chap. 3). Illumination issues are discussed in the next section.

1.5.4 *Light Sources*

Photochemistry is one of the main driving forces for atmospheric processes, so that whilst there are important dark reactions such as ozonolysis or nitrate radical (NO₃) initiated chemistry, light is required for most experiments.

The most obvious source, particularly if atmosphere-like conditions are simulated, is solar radiation and for large chambers such as Helios, EUPHORE and SAPHIR it is the only feasible option. Certain small/medium sized Teflon type chambers can be operated with either solar or artificial radiation.

The transmission of solar radiation by Teflon is good over the entire solar spectrum. Spectral radiometers inside the chamber can be used to measure the actinic flux (see also Sect. 2.3), both of the incoming solar radiation and of light reflected/emitted by the chamber floor. The disadvantage of outdoor chambers using sunlight is that experiments are dependent on the weather, because large chambers made of Teflon cannot be operated in windy conditions. Like in the atmosphere, the radiation field in the chamber changes over the course of a day-long experiment, both due to the change of the solar zenith angle and also due to short-term, transient variations caused by clouds.

Artificial radiation is used for a majority of smaller Teflon chambers and all glass and metal chambers. Depending on the main purpose of the chamber, light with a broad radiation distribution, including simulation of the solar spectrum, can be used or alternatively lamps with narrow outputs for example in the UV region (e.g. mercury lamps with emission lines at 254, 308, 365 nm) can be used. For many chambers it is possible to swap between different types of lamps.

For Teflon chambers lamps are often mounted on one side and the bank of lamps is directed into the chamber. The often cuboid nature of such chambers makes it easy to establish a uniform radiation field across the chamber. For glass chambers banks

Fig. 1.12 QUAREC chamber, Wuppertal, the lights are mounted outside the chamber providing a uniform radiation field in the actual chamber



of tubular lamps surround the cylindrical chamber. Carefully arranged, the radiation field inside the chamber can be very uniform.

The chamber construction determines the UV cut-off wavelength for example quartz is transmissive for wavelengths higher than ~ 200 nm. Arranging lamps around the chamber such that a uniform radiation field is obtained is clearly not possible for a metal chamber. Two approaches are typically used. For example in the CESAM chamber, radiations from xenon arc lamps are directed into the chamber through windows, whereas in the HIRAC chamber quartz tubes mounted inside the chamber are used as a light source (Figs. 1.12, 1.13, 1.14, and 1.15). Radiation fields in these chambers are less uniform; variations can be measured with a spectral radiometer (Sect. 2.3) and instruments can be designed to sample from various locations to test for significant spatial variations of trace gas and radical concentrations.

1.5.5 Instrumentation

The type of instruments installed at the chamber depends on the primary purpose of the individual study, for example, aerosol and gas phase experiments will require different measurements. Table 1.5 summarizes typical instrumentation and measurement approaches utilized in chambers. Table 1.5 is structured into groups of instruments according to measurement parameters. Specialized and custom-built instrumentation may require significant technical support to ensure their operation. In some cases, high costs for commercial systems can balance low, long-term running costs.

It is important to consider what instrumentation is going to be applied to the chamber in advance of the construction, e.g. to allow for sufficient space and air conditioning. Although most commercial instruments and equipment that take samples for later offline analysis can be easily placed at the chamber, some components that are directly attached to the chamber (e.g. mirrors used for FTIR spectroscopy or special

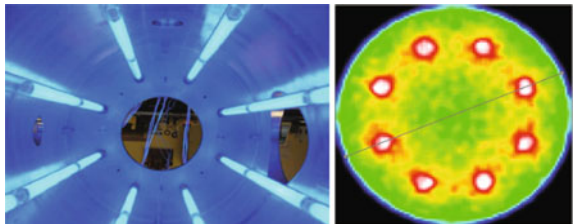
Fig. 1.13 CESAM Chamber, LISA, the chamber is illuminated from above



Fig. 1.14 FORTH chamber, a Teflon chamber with side wall illumination



Fig. 1.15 HIRAC chamber showing internal illumination and modelling of resultant radiation field across the chamber. Reproduced from Seakins (2010)



cavity ring-down systems) have to be considered in the early planning of the chamber construction. Mirrors need to be mounted where they are unaffected by vibrations from fans or pumps and the mounting needs to be rigid with respect to changes in pressure or they need to be easily adjustable. Purge gas flows may be needed for

Table 1.5 Summary of typical instrumentation and measurement approaches utilized in chambers, structured in order to group the instrumentation according to measurement parameter

<i>Gas phase concentration</i>			
Parameter	Instrument type	Online/offline/in situ	References
NO concentration	Chemiluminescence NO _x analyser Mo converter	Online	Dunlea et al. (2007)
NO and NO ₂ concentration	Chemiluminescence NO _x analyser blue light Converter	Online	NO ₂ : Ryerson et al. (2000)
NO ₂ concentration	NO ₂ cavity-based absorption spectroscopy	Online	Fuchs et al. (2010), Kebabian et al. (2005)
Ozone concentration	UV absorption O ₃ Analyser,	Online	Parrish and Fehsenfeld (2000)
Ozone concentration	Chemiluminescence	Online	Ridley et al. (1992)
SO ₂ concentration	fluorescence SO ₂ Analyser	Online	Krechmer et al. (2016), Krechmer et al. (2017)
CO ₂ , CO, H ₂ O, CH ₄ concentration	CRDS	Online	https://www.picarro.com/products/g2401_gas_concentration_analyzer
HCHO	Hantzsch reaction	Online	Junkermann and Burger (2006)
HONO	Chemical derivatization; HPLC or photometer absorption	Online	Kleffmann et al. (2002), Afif et al. (2016)
VOC, OVOC concentration	PTR-MS, CIMS	Online	DeGouw and Warneke (2007), Taipale et al. (2008), Barber et al. (2012), Cappellin et al. (2012), Duncianu et al. (2017)
VOC concentration	Sorbent cartridges + GC-MS	Offline	Rossignol et al. (2012a, b)
VOC concentration	Automatic GC- FID	Online	De Blas et al. (2011)
VOC, NO _x , NO _y , Ozone	long path FTIR	In situ	Barnes et al. (1985), Doussin et al. (1997)

(continued)

Table 1.5 (continued)

OH Reactivity	Flash-photolysis combined with laser-induced fluorescence Comparative Reactivity Method CRM	Online	Fuchs et al. (2017) and references therein
NO ₃ Reactivity	CRDS	Online	Dewald et al. (2020)
Water mixing ratio	Hygrometer gage, chilled mirror	In situ	Fahy et al. (2014)
Relative humidity	polymer sensors	In situ	Fahy et al. (2014)
<i>Chamber physical properties</i>			
Parameter	Instrument type	Online/offline/in situ	References
Temperature	Thermocouple resistance thermometer	in situ	Dias et al. (2017)
Pressure	Capacitance manometer	in situ	Blado et al. (1970)
Irradiation spectrum/actinic flux	spectral radiometer	in situ	Bohn et al. (2008)
<i>Radical measurements</i>			
Parameter	Instrument type	Online/offline/in situ	References
OH	Long Path DOAS Laser-induced fluorescence Chemical-ionization mass spectrometry	Online, in situ	Schlosser et al. (2009)
OH	Faraday Rotation Spectroscopy	Online	Zhao et al. (2018)

(continued)

Table 1.5 (continued)

HO ₂	NIR-CRDS	Online, in situ	Onel et al. (2017)
HO ₂	Chemical conversion and Laser-induced fluorescence	Online	Fuchs et al. (2010)
HO ₂	CIMS(Br-)	Online	Albrecht et al. (2019)
CH ₃ O ₂	NIR-CRDS	Online, in situ	Onel et al. (2020)
CH ₃ O ₂	Laser-induced fluorescence	Online	Onel et al. (2017)
RO ₂	CIMS	Online	Nozière and Hanson (2017)
RO ₂	ROx-LIF	Online	Fuchs et al. (2008), Whalley et al. (2018)
Rox	ROx-MAS, PerCIMS	Online	Hanke et al. (2002), Edwards et al. (2003)
Rox	PERCA	Online	Cantrell et al. (1984)
Hydroperoxides, peroxides	HPLC with FLD detector	Offline	Lazrus et al. (1986)
NO ₃	DOAS, cavity-based absorption spectroscopy	Online	Dorn et al. (2013), Fouquateau et al. (2020)
N ₂ O ₅	FTIR, cavity-based absorption spectroscopy		Fuchs et al. (2012a, b)
<i>Particulate matter physical properties</i>			
Parameter	Instrument type	Online/offline/in situ	References
Total concentration/Size distribution	Particle Size Magnifier (PSM)	Online	Vanhanen et al. (2011)
Total concentration	Diethylene Glycol-Condensation Particle Counter (DEG CPC)	Online	Jiang et al. (2011), Wimmer et al. (2013)

(continued)

Table 1.5 (continued)

Size distribution, Ion size distribution	Neutral and Air Ion Spectrometer (NAIS)	Online	Mirme and Mirme (2013)
Ion size distribution	Cluster Ion Counter (CIC)	Online	
Ion size distribution	Air Ion Spectrometer	Online	Mirme et al. (2007)
Total concentration	Condensation Particle Counter (CPC)	Online	Stolzenburg and McMurry (1991), Hering et al. (2005)
Total concentration/Size distribution	Condensation Particle Counter Battery (CPCb)	Online	Kulmala et al. (2007)
Size distribution	Differential Mobility Analyzer Train (DMA train)	Online	Stolzenburg et al. (2017)
Size distribution	Scanning Mobility Particle Sizer (SMPS/MPSS)	Online	Wang and Flagan (1990), Gaie-Levrel et al. (2020)
Size distribution	OPC–single wavelength (eg. Grimm)	Online	Heim et al. (2008)
Size distribution	OPC–white light	Online	Heim et al. (2008)
Size distribution	APS	Online	Pfeifer et al. (2016)
Aerosol spectral absorption	Photoacoustic spectrometer	Online	Arnott et al. (1999)
Aerosol spectral absorption	Aethalometer	Online	Arnott et al. (2005), Di Biagio et al. (2017a, b)
Aerosol spectral scattering	Nephelometer	Online	Andersoin and Ogren (1998)
Aerosol spectral extinction/scattering/SSA	External CRDS–Optical cavities	Online	Massoli et al. (2010)

(continued)

Table 1.5 (continued)

Aerosol spectral absorption	Filter sampling–Multi-Wavelength Absorbance Analyzer (MWAA)	Offline	Massabò et al. (2018)
Particle morphology/shape distribution	filter sampling–Electron microscopy	Offline	Chou et al. (2006)
Mass concentration	Online tapered element oscillating microbalance (TEOM)	Online	Tortajada-Genaro and Borrás (2011), Roberts and Nenes (2005)
Aerosol hygroscopicity	Hygroscopic Tandem Differential Mobility Analyser (HTDMA)	Online	Denjean et al. (2014), Good et al. (2010)
Cloud condensation Nucleous counter	CCN counter	Online	Wex et al. (2009)
Ice Nucleous Counter	IN counter	Online	DeMott et al. (2018), Mölher et al. (2021)
<i>Particulate matter concentration and composition</i>			
Parameter	Instrument type	Online/ offline/in situ	References
Organic aerosol chemical composition	Filter sampling - derivatization - GC-MS	Offline	Chiappini et al. (2006)
Elemental carbon and organic fraction of particle	EC/OC (Sunset)	Offline	Massabò et al. (2016)
Aerosol composition	ACSM-ToF Aerodyne		Ng et al. (2011a, b)
size segregated Aerosol composition	AMS		Kiendler-Scharr et al. (2009a, b)

(continued)

Table 1.5 (continued)

Surface state and composition	Filter sampling–X–ray induced photoelectron spectroscopy (XPS)	Offline	Denjean et al. (2015)
Refractory black carbon	Single Particle Soot Photometer (SP2)	Online	Laborde et al. (2012)
Inorganic/organic soluble fraction	Particle into Liquid Sampler (PILS)	Online	Pereira et al. (2015), Behera and Sharma (2012)
Inorganic soluble fraction	Filter sampling–Ion Chromatography (IC)	Offline	Chen and Jang (2012)
Organic aerosol chemical composition	filter sampling–UPLC-QTOF-MS	Offline	Nizkorodov et al. (2011), Hamilton et al. (2008)

optical systems to keep mirrors clean. Access to important equipment that may need regular cleaning or service must be assured. Some instruments will extract significant volumes of gas posing requirements on the chamber volume to ensure that dilution does not become a major loss term. A mechanism of regulating the replenishment flow to maintain a certain pressure or volume may be required.

There is a very strong synergy between chamber and field communities in terms of instrumentation, with chambers being used to design, develop, validate and compare field instruments. In general, instruments that work well in the field will be suitably sensitive and robust to ensure efficient use within chambers.

Homogeneous mixing within the chamber has to be ensured. This can be tested through a comparison of measurements that derive average concentrations across the pathlength of the system and point measurements at a single location, as well as through sampling from different locations. In addition, careful design of sampling systems (material, residence time, heating) and location of the instrumentation to minimize transfer distance limits the effect of sampling losses or transformation of reactive or instable species during the sampling process.

Making sensitive measurements of complex systems is a challenging task and even if carefully operated, systematic errors or inferences can occur. Having multiple, complementary methods (or regularly participating in inter-comparisons) can help identify these problems.

The above discussion focused on how scientific objectives and considerations influence the chamber design, construction and instrumentation. This section can only give a brief outline on considerations. This section can be used as an overall introduction, but details can be found in technical papers and reports. There is no perfect chamber design; each system has its own advantages in meeting particular objectives, but also disadvantages. In fact, having a variety of chamber designs and performing comparisons (i.e. reference experiments as detailed in Sects. 2.4 and 2.5, Donahue et al. 2012) highlights issues that would easily be missed in standardized approaches.

1.6 Conclusion

The original use of “smog chambers” for investigating chemical transformations in the atmosphere, for quantification of the rate, extent and relevance of the various possible pathways, for the identification of secondary pollutants remains just as relevant today as it did many decades ago. Indeed, the models that utilise chamber-derived data are still far from explicit, i.e. they do not include all of the processes that are required to represent and forecast the actual atmospheric composition, and there is still room for improvement, as well as the possibility of incorporating new chemistry to address future challenges. At the same time, the field of experimental atmospheric simulations has been extremely active over 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. 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 allow a much broader range of scientific and societal needs to be addressed, including the direct and indirect climate effect of atmospheric pollutants, the impact of air composition on health and cultural heritage, as well as on the various compartments of the Earth system. The application of simulation chambers in some of these areas is still in the early stages, but rapid progress is being made and already producing data that will help to open new ways of considering the complex interplays between atmospheric transformation and impacts.

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