Opinion: The strength of long-term comprehensive observations to meet multiple grand challenges at different environments and in the atmosphere

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Abstract. To be able to meet global grand challenges (climate change; biodiversity loss; environmental pollution; scarcity of water, food and energy supplies; acidification; deforestation; chemicalization; pandemics), which all are closely interlinked with each other, we need comprehensive open data with proper metadata, along with open science. The large data sets from ground-base in situ observations, ground and satellite remote sensing and multiscale modelling need to be utilized seamlessly. In this opinion paper, we demonstrate the power of~~describe~~ the SMEAR (Station for Measuring Earth surface – Atmosphere Relations) concept ~~We also demonstrate its power~~ via several examples, such as detection of new particle formation and their subsequent growth, quantifying atmosphere-ecosystem feedback loops, combining comprehensive observations with emergency science and services, as well as studying the effect of COVID restrictions on different air quality and climate variables. The future needs and the potential of comprehensive observations of the environment are summarized.

1 Background

The Earth is facing several environmental challenges on a global scale, often called "Grand Challenges" (https://www.wcrp-climate.org/grand-challenges/). The growing population (https://pdp.unfpa.org) needs fresh air, fresh water, food and energy,

while at the same time climate is changing, many cities have challenges with air quality, biodiversity is decreasing and supplies of fresh water, food and energy are diminishing. Since these Grand Challenges are highly connected and interlinked, not only with each other but also with e.g. pandemics, they cannot be solved separately since potential solutions are tightly coupled with each other (e.g. Kulmala et al., 2015; Lappalainen et al., 2016; Nolan et al., 2018; Laughner et al., 2021; Wang et al., 2023). However, the solutions may also include unexpected trade-offs (e.g. Fastre et al 2020). Therefore, integrated, comprehensive, big open data are required (Kulmala et al., 2021), together with a research and innovation framework in which a multidisciplinary research with critical mass of scientists utilising proper resources is connected to fast-tracked policy making and wide stakeholder community. This allows aiming for practical solutions based on deep scientific understanding.

The global challenges are intimately linked to interactions and feedbacks between the different compartments of the planet Earth at different spatial and temporal scales. Fundamentally, the atmosphere is closely interconnected with various other parts of the Earth, including biosphere, hydrosphere, cryosphere and lithosphere as well as urban surfaces over a range of time and spatial scales varying from seconds to millennia (Wanner et al., 2008). The sources, sinks and atmospheric concentrations of reactive trace gases, greenhouse gases and aerosol particles depend strongly on each other via physical, chemical and biological processes (e.g. Arneth et al., 2010, Stocker et al., 2013; Kulmala et al., 2014a; Unger, 2014, Green et al., 2017; Smith et al., 2023). Furthermore, both human actions and natural feedback mechanisms between the biosphere and atmosphere have substantial impacts on interactions between these atmospheric constituents and their influences on air quality and climate (Raes et al., 2010; Stocker et al., 2013; Kulmala et al., 2015; Kulmala, 2015; Nolan et al., 2018; Doherty et al., 2022; Wang et al., 2023). The importance of atmospheric aerosol particles on climate and human health in both regional and global scales has attracted a plenty of research interest during the recent years (Butt et al., 2017; Boy et al., 2019; Bellouin et al., 2020; Lappalainen et al., 2022; Lintunen et al., 2023). Despite these efforts, atmospheric aerosol particles remain perhaps the least known factor influencing radiative forcing, causing thereby large uncertainties in predicting the future behavior of the climate system (IPCC 2013, 2021).

The global annual cost of climate change impacts are estimated to reach hundreds of billions of euros by 2030 (UNEP, 2016; Köberle et al., 2021) and, with an increasing global warming, this cost is expected to increase strongly in the future. According to the World Economic Forum, climate change has cost the EU €145 billion in a decade (https://www.weforum.org/agenda/2022/12/climate-europe-gdp-emissions/). There is an urgent need for improving climate projections, reducing greenhouse gas emissions and developing options to sequester terrestrial carbon by simultaneously taking into account the other climate forcers, such as atmospheric aerosol particles and trace gases. In addition, in order to have better understandings of natural and anthropogenic sources and sinks of carbon and of atmospheric processes influencing air quality, we need to develop existing monitoring and forecasting systems of the terrestrial carbon cycle, and to both enhance and improve measurements from process levels to a global scale. These practical needs provide emerging business opportunities for various industries. For example, European Green Deal Investment will mobilize at least one trillion euros of sustainable

investments over the next decade (COM 2020). The information produced by using new verification systems is essential for society to design economically and socially optimal sustainability strategies and climate-neutrality pathways and to be able to meet Paris Agreement targets (Kriegler et al., 2018). The importance of comprehensive and standardized measurements is also underlined by rapid development of the environmental data analysis based on artificial intelligence (AI). The quality of the AI based environmental analysis is as good as the measured source data. The novel Earth Virtualization Engines (EVE) concept (https://eve4climate.org/) in under development and the foreseen digital infrastructure of multi-tiered climate information for various type of users would build on optimal earth-system data integration and monitoring by using AI methods like machine learning (Stevens et al. 2023).

Open science and open data are essential. Open science, the sharing of knowledge and data as early as possible in the research process (Vicente-Saez and Martinez-Fuentes, 2018) are essential as addressed by bodies like European Commission(European Commission, 2021). Such in-situ data can be obtained from global networks, such as GAW (Global Atmospheric Watch) and FluxNet (Smith et al. 2012; Baldocci, 2019), and continental-scale infrastructures such as TERN (Terrestrial Ecosystem Research Network in Australia), NEON (National Ecological Observatory Network in US), ASCENT (Atmospheric Science and Chemistry mEasurement NeTwork in US), AmeriFlux (Boden et al. 2016), ChinaFlux (Yu et al. 2006) AsiaFlux (Mizoguchi et al. 2008), AfriFlux (Ciais et al. 2011), and the European RI's ICOS (Integrated Carbon Observation System), ACTRIS (Aerosols, Clouds, and Trace gases Research Infrastructure), and eLTER (Integrated European Long-Term Ecosystem, Critical Zone & Socio-Ecological Research Infrastructure) (Loescher et al., 2022). However, often the in -situ data is covering only some components of the Earth system and the proper integration of different datasets is lacking. This can be overcome by co-locating different measurements, which enables new knowledge of the interactions and feedbacks between the Earth components (biosphere, hydrosphere, atmosphere, and geosphere), and allows for science-based solutions related to the interlinked Grand Challenges. An example of co-located measurments at a single station enabling new knowledge of the interactions and feedbacks between the Earth components, spearheading science-based solutions related to the interlinked Grand Challenges, is Station(s) for Measuring Earth surface – Atmosphere Relations (SMEAR).

The primary objective of this paper is, by using a few examples based mainly on data from the SMEAR II station located in Hyytiälä, Finland, to demonstrate the power of comprehensive, continuous and integrated long-term observations in a way toward addressing some of the Grand Challenge discussed above. This kind of an approach is closely tied with the SMEAR concept introduced earlier and summarized briefly in the next section. Besides research associated with the Grand Challenging, we demonstrate the strength of the SMEAR concept when rapid and unpected changes occur in the environment and atmosphere.

2 SMEAR concept

The SMEAR concept is based on comprehensive, continuous, and integrated long-term observations (Hari and Kulmala, 2005; 115 Hari et al., 2016). It has been developed to meet Environmental Grand Challenges and to collect big open data sets in order to test theories and develop models at the interfaces of different Earth components. Such unique environmental open data can contribute to solving burning questions of society – even questions that are currently unforeseen.

Current observations (see IPCC 2013, 2021) are fragmented, which means that typically different infrastructures are measuring 120 greenhouse gases, aerosols, air quality, ecosystems, climate and biodiversity. These measurements are conducted in different locations and environments, and often during relatively short campaigns. However, to meet the ongoing environmental challenges, an integrated approach with long-term measurements is needed. In practice this means co-location of various infrastructures, which enables simultaneous measurements of different Earth components (Guo, 2018; Kulmala et al., 2021; Lintunen et al., 2023). Changes in one of these components are directly or indirectly communicated to the others via intricately 125 linked processes and feedbacks occurring at their interfaces (e.g. Stocker et al., 2013; Nolan et al., 2018; Smith et al., 2023).

The SMEAR stations, together with the SMEAR concept, were established before the various international environmental research infrastructures were established (Hari and Kulmala, 2005). Today the four Finnish SMEAR stations and international SMEAR-like stations (Kulmala et al., 2021) are contributing to the European ESFRIs focused on standardized measurements 130 on atmospheric composition and ecosystem processes like ICOS, ACTRIS, eLTER and AnaEE. SMEAR II is participating to several global Earth Observation systems and networks such as WMO GAW and/or EMEP, GEO-GEOSS, FluxNet,

AERONET and SolRad-Net. The key aspect is the co-location of these different thematic networks into a single site. This allows multi- and interdisciplinarity open science based on the collected data.

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Crucial in the SMEAR concept is that it measures a set of variables needed for the atmosphere-Earth surface interactions and feedback analysis, providing fundamental knowledge for the climate change mitagiation and adapation plans. The Earth

145 surface can be, for example forest, lake, ocean, peatland, urban area, glacier, agricultural land, etc. The most established station, SMEAR II, is located in Hyytiälä, Finland, and it includes measurements of over 1200 different variables (Kulmala et al., 2021). The measurements are conducted at different scales from small chamber enclosures to a regional scale, which is made possible by the 128-m-high measurement tower at the SMEAR II station. The measurements include meteorological variables, atmospheric compositions and fluxes (aerosols, clouds, atmospheric chemistry, greenhouse gases etc.), as well as variables

150 describing ecosystem functioning and soil dynamics. Long-term *in situ* measurements are accompanied by remote sensing, experiments (both lab and field) and multi-scale modelling. Such an approach enables us to tract the regional and long-range transboundary pollution transport and to evaluate e.g. trends in measured concentrations and fluxes, process dynamics, and feedbacks between processes and Earth components, such as soil-forest-atmosphere and forest-soil-streams-lake.

155 An important part of the SMEAR concept is open access to the research infrastructure and open data (https://smear.avaa.csc.fi/). These data are massive and heterogeneous, and thus challenging to manage, but the easy access to these data and both harmonised and standardised ways to analyse it are important (Junninen et al., 2009). As a summary, we can state that we need to work towards an open, integrated approach that can be accomplished with a global SMEAR network, a global Earth observatory (Hari et al., 2016; Kulmala 2018).

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In the following section, we give examples of capabilities of the SMEAR concept addressing different kinds of research questions: atmospheric new particle formation, COBACC feedback loop that combines terrestrial carbon sink to aerosol source, COVID impacts on air quality, and long-term trends in some of the quantities essential for air quality and climate research. We also discuss the future role of comprehensive measurements to detect unexpected changes in the environment

165 and atmosphere.

3 The utilization of comprehensive data sets

Here we provide examples on how we have used comprehensive data sets to meet several scientific and societal challenges and discuss briefly what has been learnt from these investigations.

3.1 Atmospheric new particle formation (NPF)

170 A well-known example of the usefulness of comprehensive, long-term observations are investigations related to atmospheric NPF (e.g., Mäkelä et al., 1997; Kulmala et al., 2013). Before such observations, more emphasis was placed on binary nucleation in the stratospheric conditions (e.g, Hamill et al., 1982). When looking at the scientific literature published prior to mid-1990´s, the common thought appears to have been that in the troposphere, NPF is a relatively rare and local phenomenon with minor contributions to regional or global aerosol particle budgets. However, the first long-term observations of particle number

175 concentrations revealed a frequent occurrence of NPF and its regional character in a boreal forest environment (Mäkelä et al., 1997), and later observations confirmed the same to be the case in many other types of atmospheric environments (e.g. Kerminen et al., 2018; Nieminen et al., 2018; Chu et al., 2019; Brean et al., 2023). Motivated by long-term observations, explicit description of NPF was then included in several large-scale modeling frameworks. Simulations using such models demonstrated that NPF is the dominant source of the particle number concentration in the global atmosphere, and an important

180 contributor to concentrations of cloud condensation nuclei (e.g. Merikanto et al., 2009; Gordon et al., 2017).

In order to understand how atmospheric NPF is connected with climate and air pollution, or emissions to the atmosphere, one needs to quantify the mechanistic pathways of atmospheric NPF and its basic characteristics, such as its frequency and associated particle formation and growth rates. Our mechanistic understanding on the initial steps of atmospheric NPF,

185 clustering, relied for a long time on theories and laboratory experiments, and with a belief that the only important clustering mechanisms in the atmosphere is the binary nucleation between sulfuric acid and water vapors (Malila, 2018). Atmospheric observations provided increasing evidences on the existence of multiple and possibly more complex clustering pathways (e.g. Kulmala et al., 2014b). Such findings inspired comprehensive and dedicated laboratory experiments in the CLOUD chamber at CERN (e.g. Kirkby et al., 2011; Almeida el al., 2013; Lehtipalo et al., 2018, He et al., 2021). Many of the clustering pathways

190 quantified in these CLOUD experiments have recently been found in atmospheric observations (e.g. Sipilä et al., 2016; Jokinen et al., 2018; Lehtipalo et al., 2018; Beck et al., 2021; Yan et al., 2021), confirming the diversity of NPF in various atmospheric environments.

Atmospheric observations have revealed large differences in the NPF characteristics between different sites, as well as between

195 different seasons at individual sites (e.g. Nieminen et al., 2018; Chu et al., 2019; Deng et al., 2020, Brean et al., 2023). At sites with multi-year observations, there appears to be a notable inter-annual variability in both frequency and intensity of NPF, and in some cases also a long-term trend has been reported (Asmi et al., 2011; Nieminen et al., 2014; Saha et al., 2018; Kalivitis

et al., 2019; Neefjes et al., 2022). Figure 1 shows the monthly medians of nucleation mode particle concentrations measured at the SMEAR II station. Over the 27-year observation period from 1996 until 2022, the monthly median concentrations decreased at a rate of –0.9%/year. The temporal variability in NPF characteristics has been ascribed to changes in meteorological conditions and aerosol precursor sources, including clear influences of reduced sulfur emissions and other air pollution control actions in Europe and North America (Hamed et al., 2010; Kyrö et al., 2014; Wang et al., 2017; Saha et al., 2018) and more recently also in China (Zhao et al., 2021; Zhu et al., 2021).

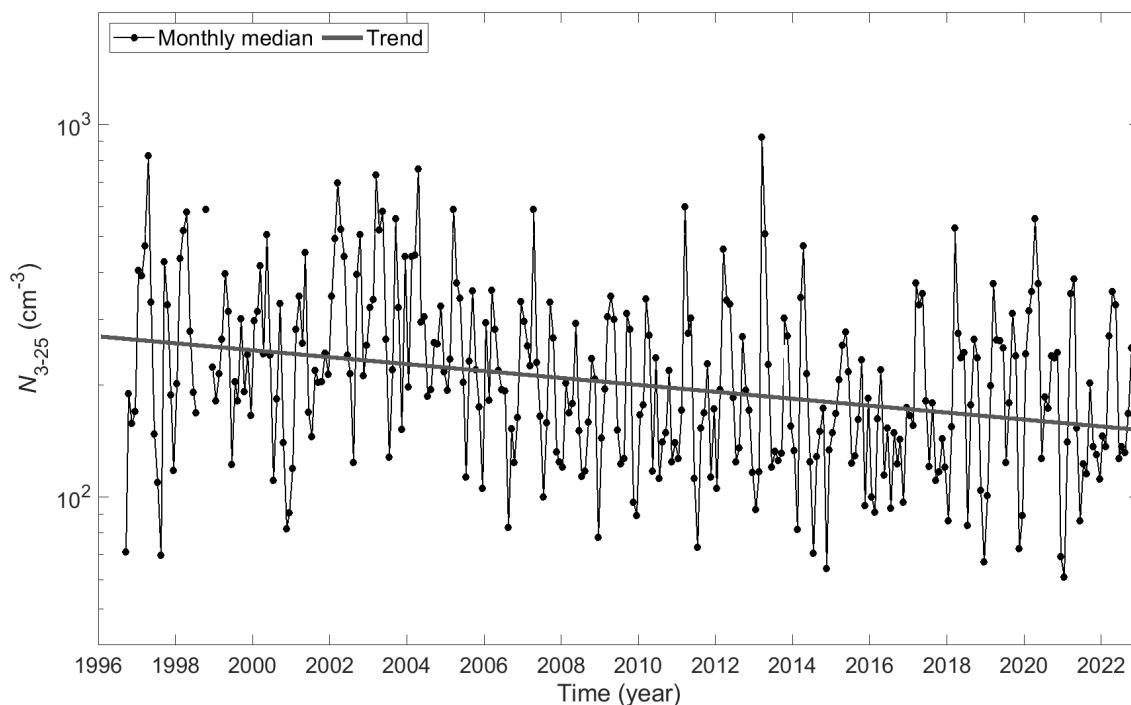



Figure 1: Monthly medians of the nucleation mode particle (3–25 nm in the mobility diameter) concentrations at the SMEAR II station in Hyytiälä, Finland. The line is a linear lin-log fit to the data and shows clearly the decreasing trend.

Long-term atmospheric observations have played a central role in atmospheric model development. The first semi-empirical parameterizations of new particle formation rates for modeling purposes were based on simultaneous and continuous measurements of gas-phase sulfuric acid concentrations and particle number size distributions (Sihto et al., 2006; Paasonen et al., 2010; Semeniuk and Dastoor, 2018). Later, long-term observations have been essential in testing the performance of large-scale models in simulating NPF and subsequent growth of newly formed particles to CCN (e.g. Spracklen et al., 2010; Fountoukis et al., 2012; Yu et al., 2015; Qi et al., 2018).

While models are likely to be the main tool for estimating the future impacts of atmospheric NPF on climate and air quality, they regularly need observations to verify their performance. In addition, many related scientific issues remain that cannot be solved without comprehensive and continuous observations. One of them is the relative importance of different clustering pathways in different environments and due to continually changing atmospheric composition in these environments. The second issue is the quantification of factors dictating the frequency and intensity of NPF, including the role of "quiet NPF", i.e. relatively weak NPF not captured by traditional NPF event analysis methods (Kulmala et al., 2022a). The third issue is to understand the growth of newly formed particles into sizes where they may act as CCN or contribute to haze formation (e.g. Ren et al., 2021; Kulmala et. 2022b; Stolzenburg et al., 2023). Related to this issue, we need long-term observations to better understand how small clusters survive while growing   larger sizes, especially in polluted environments (Kulmala et al., 2017; Tuovinen et al., 2022), in order to find out the relative importance of condensation and heterogeneous reactions in growth, and to quantify the most important precursor vapors causing this growth.

3.2 COBACC ~~and other~~ feedback loop~~s~~

To understand better the complex feedbacks between the atmosphere and ecosystems, we have developed a concept called the Continental Biosphere-Atmosphere-Clouds-Climate (COBACC) feedback loop (Kulmala et al., 2013). It utilises a multidisciplinary and integrated approach to quantify the feedbacks. The loop consists of several interrelated processes (~~Fig. 2,~~ for detailed description see Kulmala et al., 2014a; Artaxo et al., 2022, Kulmala et al., 2023): 1) the atmospheric temperature and CO_2 influences on biogenic volatile organic compound (BVOC) emissions, 2) the influence of BVOCs on the formation and growth of aerosol particles, 3) the effect of aerosol particles on clouds, 4) the effect of aerosol particles and clouds on solar radiation, in particular, on its diffuse fraction, and 5) the link between diffuse radiation and photosynthesis, and carbon sink in general.

The various processes involved in the interactions within the COBACC feedback loop occur over different time scales. Increases in atmospheric temperatures and carbon dioxide concentrations occur at inter-annual time scales, carbon cycling including photosynthesis and emission of BVOCs vary on scales from sub-hourly to seasonal, while the cloud variability and its effect on radiation that drive photosynthesis operate at sub-hourly time scales. Therefore, continuous and comprehensive observations, together with modelling, are key to solving such complex questions. In what follows, we summarize our current understanding of the feedback loop in the boreal zone (for a more comprehensive review see Artaxo et al., 2022), and indicate future directions. Continuous observations serve as a base for most of the studies cited below, and the SMEAR II data set, used in most of them, remains the most comprehensive one up to date.

Paasonen et al. (2013) considered the effect of warming climate on BVOC emissions and associated increases in >100 nm aerosol particle number concentrations (a proxy for CCN), quantifying the potential cooling effect due to this feedback. The

250 pilot study of Kulmala et al. (2014a) made the first estimate of the COBACC feedback loop using SMEAR II data, focusing on the direct aerosol effect and excluding clouds from the consideration. After that, Ezhova et al. (2018) refined and extended the analysis of the aerosol-diffuse radiation-photosynthesis part of the feedback loop using data from five sites in the boreal zone, also excluding clouds. Clouds were included in the Earth System Model (ESM) studies on the feedback loop (Rap et al., 2018; Sporre et al., 2019). However, the link between BVOC, aerosol particles and clouds in various ESMs is a source of

255 substantial discrepancies, even of different sign, in the radiation – the main driver of photosynthesis (Sporre et al., 2020). Therefore, observations remain an extremely relevant source of data for this complex question.

Based on the COBACC feedback loop, Kulmala et al. (2020) developed the CarbonSink+ concept, which, beside aerosols, takes into account the effect of forest on clouds and surface albedo. The next step is to include all radiative forcers. Current

260 COBACC feedback loop studies are directed towards quantifying the role of clouds (Fig. 2), including their interaction with the surface-based parameters and their effects on radiation and photosynthesis, based on observations. The combination of on-site and satellite observations was employed to show that clouds become optically thicker in a warmer climate with larger amounts of organic aerosol particles (Yli-Juuti et al., 2021). Furthermore, Petäjä et al. (2022) showed that continuous interaction of an air mass with emissions from the boreal forest changes the properties of this air mass over a time period of

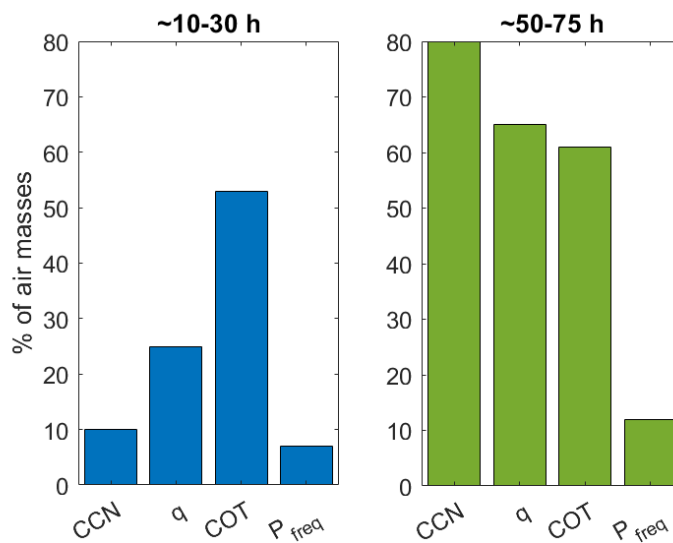
265 several days, including both aerosol physical and chemical characteristics and humidity. Both factors are important for the formation and evolution of clouds. Räty et al. (2023) extended this approach to a data set covering more than a decade and confirmed the main conclusions showing an increase in, e.g., cloud condensation nuclei, specific humidity as well as cloud optical thickness and precipitation frequency (Fig. 2).

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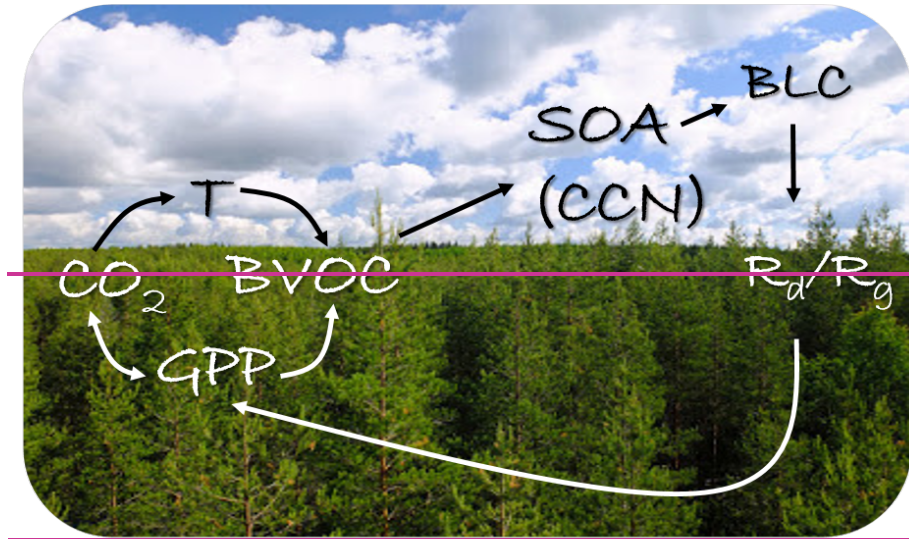
Figure 2: Illustration of forest-boundary layer clouds link (Räty et al., 2023): fraction of air masses with the parameter value above its median after 10-30 h and 50-75 h interaction with boreal forest. Parameters are cloud condensation nuclei at 0.2%

However, the outcome from this study regarding the effect of forest on cloud properties remains somewhat obscure~~. : c~~Cloud properties were taken from the satellite data sets, which drastically decreases the number of data available for analysis. To overcome this problem, e.g. the cloud classification algorithm by Ylivinkka et al. (2020) can be used. The algorithm allows quantifying some cloud properties, e.g. optical thickness for some types of clouds, whereas cloud fraction is linked to patchiness. The radiation measurements, an input parameter for this algorithm, have been measured at SMEAR II for more than two decades, and therefore the cloud-related data set can potentially be extended significantly. Overall, continuous comprehensive observations play a key role in tackling multidisciplinary problems with multiple time scales.

While the COBACC feedback has, until now, been studied primarily in the boreal ecosystem, more data to constrain similar feedbacks within other ecosystems – particularly tropical and (semi)arid as well as urban – are urgently needed.

BVOC and semi volatile organic compounds (SVOC) are closely linked to SOA formation as a function of ecosystems. Therefore, quantification and research of the fluxes of these VOCs is crucial. Field measurements of fluxes of BVOC and their oxidation products exhibiting reduced volatility, such as SVOC are challenging, and they can only be measured with rather short inlets to avoid wall losses during sampling. Recently a PTR3 instrument was used on top of the SMEAR II tower in Hyytiälä (Fischer et al., 2021) at 36 m above ground level and 15 m above the canopy of a forested ecosystem dominated by

terpenoid emitters. The PTR3 instrument was installed approximately 4 m away from the tower structure and the virtually wall-less inlet was successfully tested allowing undisturbed gas sampling from this distance. For the first time emission fluxes of sesquiterpene ozonolysis products and diterpenes were recorded. With the low flux signal-to-noise ratio achieved with the new instrumentation, we can now track and study clear diurnal patterns, even for the smallest emissions rates virtually in real time. Such intensive campaigns demonstrate the feasibility of new technology to be integrated in Flagship stations providing an extended parameter set in the future.

3.3 COVID restrictions

By the end of 2021, the global spread of COVID-19 caused by the SARS-CoV-2 virus has resulted in the loss of over 10 million lives (Adam, 2022; Msemburi et al., 2023). In China, national interventions were implemented starting from January 2020 to prevent the spread of the virus (China NHC, 2020). The strict lockdown measures associated with the COVID-19 pandemic provide a unique opportunity to investigate, in a real-world atmospheric laboratory, the direct and indirect effects of reduced emissions, as well as atmospheric chemistry and interacting processes associated with these emission changes, on air quality (e.g. Kroll et al., 2020; Jiang et al., 2021; Wang et al., 2021; Sokhi et al., 2021; Amouei Torkmahalleh et al., 2021).

This unique opportunity is a good demonstration of the strength of the SMEAR concept applied to atmospheric observations. First, in such an unplanned situation where new research activities have also been restricted, it is impossible to organize and carry out targeted intensive observations. Second, although there are several functioning observing stations, the relatively poor measurement capacity in most of them is unable to support the in-depth analyses needed for new scientific insights. To date, there are hundreds of atmospheric science studies relevant to the COVID-19 lockdown (https://docs.google.com/document/d/1UTQvW_OytC37IatMNR5qJK7qKfSylNpI2fT3pdteVZA/edit). However, a large proportion of these studies only report variations of a few atmospheric parameters and are far from providing a mechanistic understanding of changes in atmospheric processes. There are also studies that use regional models to understand the atmospheric processes during the lockdown, but these modeling results have limited verification due to the lack of comprehensive observations.

The Aerosol and Haze Laboratory of Beijing University of Chemical Technology (AHL/BUCT; Liu et al., 2020) is one of the stations fully implementing the SMEAR concept. This station was established in January 2018, and since then it has been operating uninterruptedly with a full measurement capacity. Our comprehensive observations showed that the lockdown caused changes of different magnitudes in various atmospheric parameters (Fig. 3). In general, most of the primary pollutants, such as NOx, SO$_2$, BC, and VOCs, showed a reduction in their abundance, but at different levels. For example, NOx was reduced by more than 50%, SO$_2$ by ~25% and VOCs only by ~15%. This suggests that emissions from different source sectors were affected differently by the lockdown. In contrast to the primary pollutants, most of the secondary pollutants showed

increased concentrations. Particulate nitrate, sulfate, ammonia, organics, and gas-phase highly oxygenated organic molecules (HOMs) increased by ~50–150%. This indicates that secondary pollution, i.e. the conversion of primary pollutants into secondary ones, became more efficient. This phenomenon is closely related to the increased oxidation capacity of the atmosphere, as indicated by the increased concentrations of OH, NO_3 and O_3.

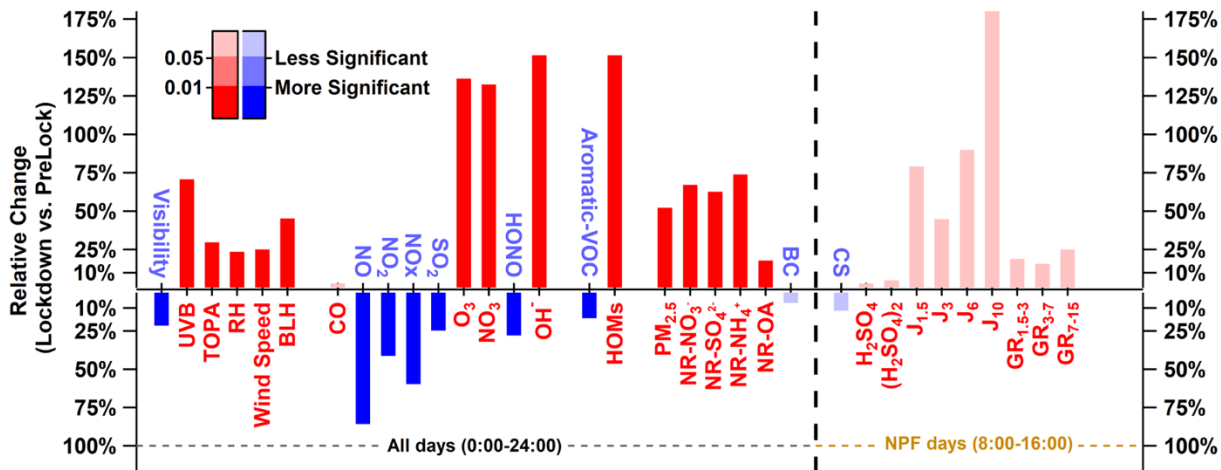



Figure 3: Variations of primary and secondary pollutants caused by the lockdown. Relative changes of atmospheric variables between the COVID-19 lockdown period (24th Jan – 5th Mar 2020) and pre-lockdown period (1st Jan – 23rd Jan 2020). The relative changes are defined as ([X]lock-[X]pre)/[X]pre × 100%, where [X] is the average of each variable. Variables associated with new particle formation (NPF) are shown only for NPF days during the daytime.

Our comprehensive data sets allow us to obtain cutting-edge knowledge in several research directions. Here, we provide two examples that provide direct observational evidence, showing the substantial influence of anthropogenic emissions on the atmospheric oxidative capacity in both daytime and nighttime.

3.3.1 How did the atmospheric new particle formation responded to COVID-19 lockdown

Yan et al. (2022) explored how NPF responded to emissions reductions in Beijing during the COVID-19 lockdown. Clustering between SA and base molecules drove the initial NPF in both the pre-lockdown and lockdown periods. Our results show that this clustering was insensitive to emission reductions. Through direct observation, this study provided evidence that traffic emissions do not appear to be a significant source of NPF in Beijing, in contrast to conclusions drawn from some recent urban studies (Rönkkö et al., 2017; Guo et al., 2020).

During the lockdown period, we hypothesized that the reduction in nitrogen oxides (NO_x) concentrations would promote particle growth. This is because NO can suppress particle growth by changing the composition of oxidized organic molecules

(OOMs) and making them more volatile on average (Yan et al., 2020). However, our study found otherwise. Although we noted changes in the composition of OOMs, especially in molecules arising from the oxidation of aromatic volatile organic compounds, there were only negligible changes in the volatility of OOMs. These results indicate that the reaction between RO_2 and NO still plays a vital role in OOM formation even after a dramatic reduction in NO_x levels. It has been suggested that the autoxidation of RO_2 will become more important in atmospheric chemistry as NO_x concentrations continue to decrease in North America (Praske et al., 2018), leading to increased toxicity of peroxide-driven particles and the formation of secondary organic aerosols (Zhao et al., 2017). However, our findings suggest that these harmful effects on human health and air quality in Beijing are less likely to arise in the immediate future.

3.3.2 Enhanced formation of secondary organic carbon associated with NO_3 radical

Carbonaceous aerosols are acknowledged to have have significant impacts on climate change, Earth's radiation balance, visibility, and human health (Donahue et al., 2009; Bond et al., 2013; IPCC, 2021). We examined carbonaceous aerosols measured with an OC/EC analyzer between 1 December 2019 and 15 March 2020 in Beijing, encompassing the COVID-19 pandemic period (Feng et al., 2022). Our findings showed that anthropogenic gas-phase pollutants and primary organic compounds were greatly reduced during the lockdown period. However, we also observed the emergence of enhanced nighttime secondary organic carbon, which we attributed to nocturnal chemistry associated with the oxidation by NO_3 radical. Our results indicate that this nocturnal chemistry phenomenon warrants greater attention in efforts to reduce PM concentration in China.

3.4 Long-term trends in comprehensive observations of atmospheric variables

The long-term observations at the SMEAR II station in Hyytiälä, Finland, cover measurements of trace gas concentrations (SO_2, O_3, NO_x, CO, CO_2) as well as volatile organic compounds (VOCs, such as monoterpenes), which are measured on multiple heights above the ground at the 128-m-high mast. The continuous time-series of measurements starting from 1996 allow us to quantify long-term trends in these variables (Fig. 4). In addition, mass spectrometer measurements of sulphuric acid (H_2SO_4, the main oxidation product of SO_2) started in 2016, and proxy calculations based on the measured H_2SO_4 concentrations enable extending this time series (Petäjä et al., 2009; Dada et al., 2020).

The monthly-median concentration of the H_2SO_4 proxy has a decreasing trend of –2.6%/year (Fig. 4a). The H_2SO_4 proxy is calculated based on the production of H_2SO_4 due to the oxidation of SO_2 by OH radicals and via stabilized Criegee intermediates which are produced in ozonolysis of monoterpenes (Dada et al., 2020), and on the loss of H_2SO_4 due to its condensation into pre-existing particles. Both the source and sink terms of H_2SO_4 have decreasing trends in Hyytiälä during 1996–2022, being –2.7%/year for the SO_2 concentration –1.0%/year for the condensation sink (Fig. 4c-d). The stronger

decrease in the H_2SO_4 precursor vapor concentration compared with the H_2SO_4 sink seems to determine the long-term trend observed in the sulphuric acid proxy concentrations.

The monoterpene concentrations are characterized by a larger year-to-year variability and do not show a statistically significant trend (Fig. 4b) (see also e.g., Tarvainen et al., 2005; Taipale et al., 2011; Rantala et al.; 2015, Hellen et al., 2018).4b). In the summertime, the monoterpene concentrations are highest during the year and have stayed relatively constant, whereas the annually lowest concentrations during winter and spring show also the largest variability between years. The long-term data show strong seasonal and diurnal patterns in emission rates, which mostly relate to changes in temperature and partly also light availability. Furthermore, vegetation phenological events and biotic and abiotic stresses produce high emission peaks (e.g., Aalto et al., 2014). The amplitude of the daily and seasonal variations in monoterpene emission rates is high and masks the potential climate change effect over the years. This emphasizes the importance of versatile, comprehensive measurements for quantifying atmospheric processes.

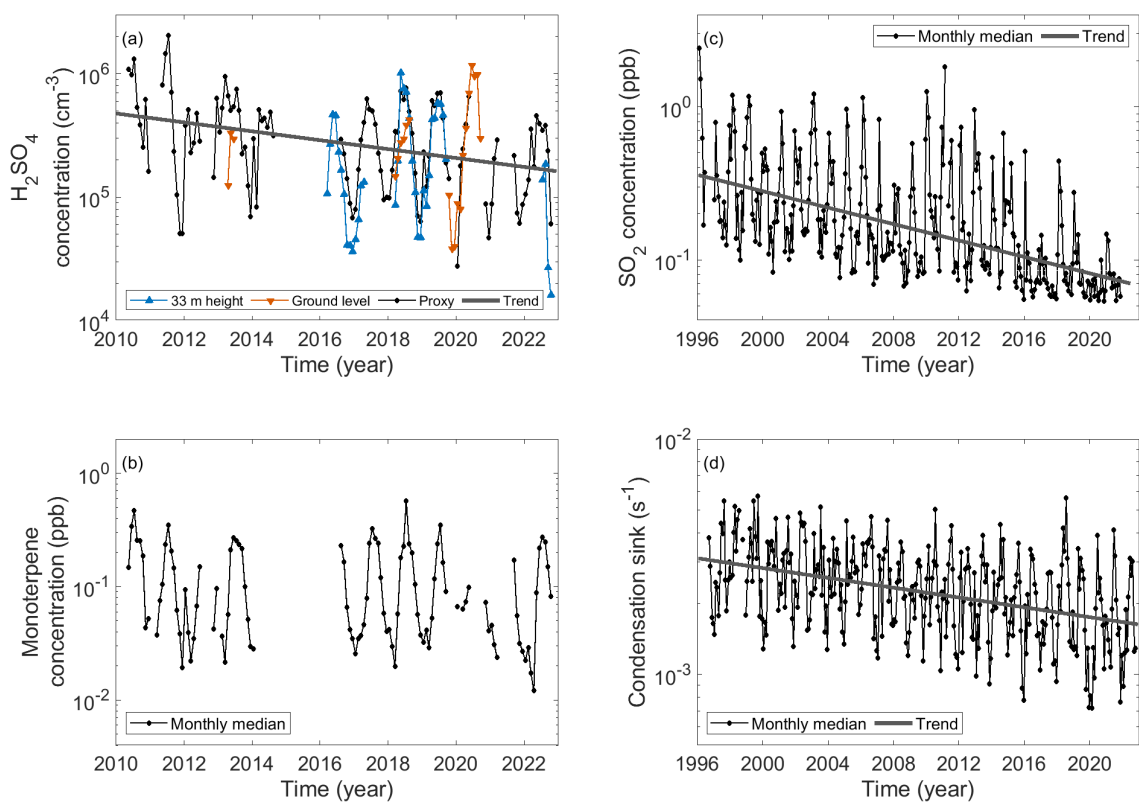



Figure 4: Trends in (a) sulphuric acid concentration, (b) monoterpene concentration, (c) sulphur dioxide concentration, and (d) condensation sink. Note the different time periods: in panels (a) and (b) 2010–2022, and in panels (c) and (d) 1996–2022. The dotted black lines show monthly medians of observations and the grey solid lines are trends fitted to the logarithmic values of the monthly data. Monoterpene concentrations do not have a statistically significant trend and therefore the trend line is not shown in panel (b).

4 Integration of the data from satellites, models and in situ observations

4.1 Satellite and Airborne observation

Satellites provide data on a global scale of atmospheric composition, radiation, surface properties and meteorology. Passive satellite measurements of atmospheric gases and aerosols are representative over an entire atmospheric column, and hence they are not directly comparable to in situ measurements. Although satellites cannot provide as detailed and wide range of different atmospheric parameters as comprehensively equipped in-situ measurement stations, such as SMEAR II, with their spatial coverage they can provide very valuable and complementary information. After understanding and analyzing in-situ and satellite measurements together on a station-by-station basis, satellite data can enable the transition from point-like measurements to the interpretation of regional variability in atmospheric processes (e.g. Viatte et al., 2021; Pseftogkas et al., 2022; Hakala et al., 2019).

One example of utilizing satellite data, when moving from pointwise to global (or regional) analysis, has been to better understand the new particle formation (NPF) phenomena on a larger spatial scale (Kulmala et al., 2011; Sundström et al., 2015). As satellites cannot essentially detect aerosol particles smaller than 100 nm in diameter, these observations as such are not directly applicable for NPF studies. However, satellites provide information on many atmospheric parameters tightly linked to NPF, such as UV radiation, trace gas concentrations and estimations on ambient aerosol loads. By analyzing these data with detailed in situ measurements, it is possible to develop merged satellite variables that can be used to study the regional variation of NPF. Other examples are various climate-related feedback loops, e.g. between the atmosphere and the biosphere, where satellite observations could have the potential of increasing the understanding of the spatial scale variation. Such development work would not be possible without SMEAR-type observations that have the capability of providing process-level understanding of the phenomena. It is also essential that such extensive in situ observations exist in various environments, so that the sensitivity of satellite observations in these kinds of applications could be properly tested.

Passive satellite instruments provide typically so-called columnar measures, and for instance aerosol optical depth (AOD) is the vertically-integrated aerosol extinction. Similarly, gas concentrations measured by satellite represent column or partial column concentrations, typically over a tropospheric column. Therefore, comparison with surface in-situ measurements cannot offer a direct validation for the measurements made by satellite instruments. However, all possible columnar measurements (e.g. AOD by ground-based sun-photometers) at the same ground station, on the other hand, would facilitate satellite data validation and provide information on the accuracy of the satellite observations. Currently, the SMEAR II station in Hyytiälä is accompanied with an Aerosol Robotic Network station (AERONET, Holben et al., 1998), which allows a direct validation of satellite-based aerosol observations. Moreover, satellite validation would strongly benefit from reliable gas and aerosol vertical profile observations from the surface level up to the stratosphere.

One possible future pathway in better bridging the spatial-scale gap between pointwise in-situ measurements and large-scale satellite measurements would be to utilize unmanned aerial vehicle (UAV) measurements (Motlagh et al., 2023). Satellite data have spatial resolutions limited to a few hundred meters at best, and in atmospheric observations more typically to kilometers. Drone measurements (e.g., Kezoudi et al., 2021) could bring more insight into the sub-kilometer scale variations if carried out in the vicinity of SMEAR-type stations at the time of the satellite overpass.

4.2 Model frameworks

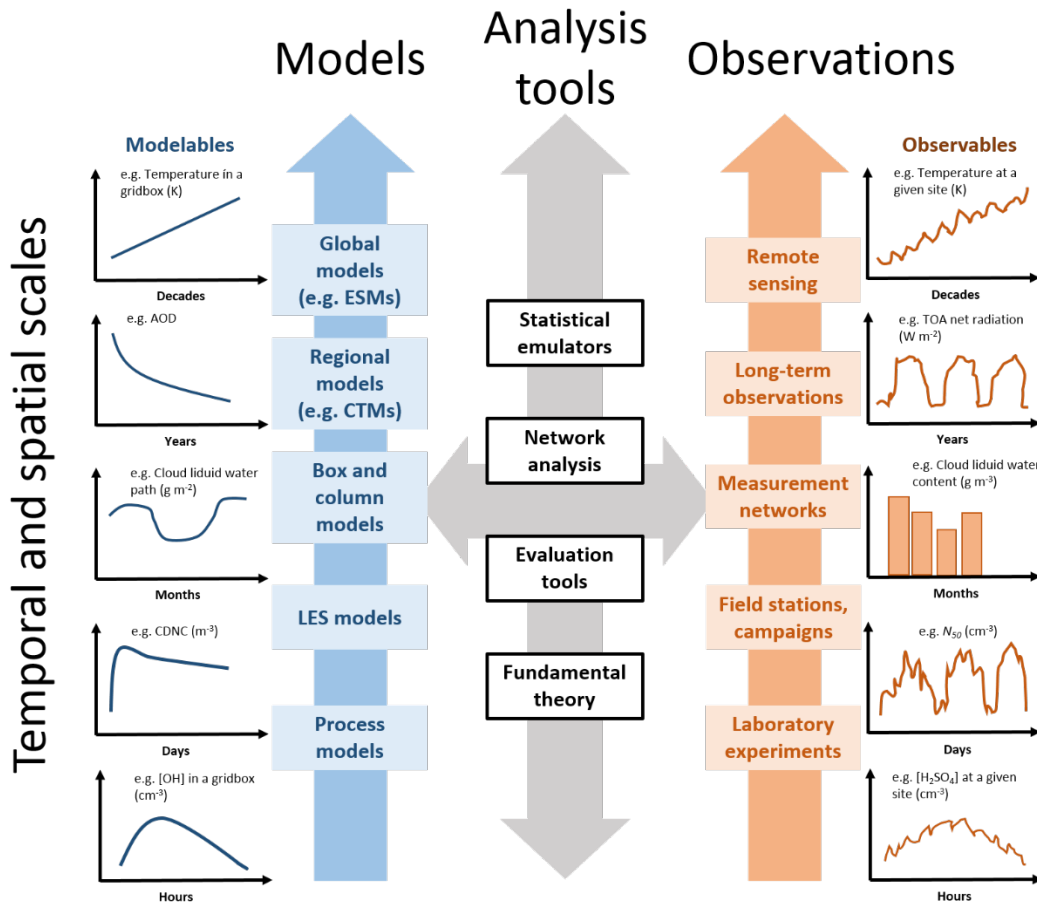
Model frameworks for the global climate and Earth systems have been constructed to replicate real-world processes and interactions as closely as necessary to understand the current state of these systems, reasons for past changes, and ultimately to simulate future climate pathways in order to support adaptation and mitigation efforts (Bauer et al., 2021). Modern Eerth System Models (ESMs) combine an increasing number of individual components, including not only the physical ocean and atmosphere models but also detailed descriptions of chemistry, aerosols and the biosphere (e.g. Döscher et al., 2022). This increase in model complexity has established groundbreaking research of Earth system feedbacks and quantification of their strength in current and future climate (Sporre et al., 2019; Thornhill et al., 2021).

Despite rigorous validation of its individual process components, evaluation and constraining of highly-coupled ESMs remains difficult due to the large number of interactions and feedbacks within the Earth system (e.g. Sporre et al., 2020). In the temporal and spatial scales of ESMs even the observational record as a whole remains brief and irregular, and therefore the observations must be extended by proxies of changes in the historical period (Wandji Nyamsi et al., 2020) and towards the Earth's deep past over millions of years (Wong et al., 2021). Only integrated long-term observations can provide multidisciplinary data to support evaluation of simulated Earth system feedbacks and their components.

With increasing complexity and process details, ESMs can arrive at correct results via wrong reasons and counteracting biases. The advancement of spatial resolution and process descriptions within ESMs already allows evaluation at the process-scale. For example, NPF events can be co-analyzed from ESMs and long-term datasets (Bergman et al., 2022). Such process-oriented analysis is essential for validating the reasons for biases or systematic errors in simulated properties (e.g. CCN), but this requires dedicated long-term observations to constrain the models throughout distinct climate states and changing environment (Fanourgakis et al., 2019). Recent advances in trajectory-based analysis of ESMs provide a novel way to investigate simulated air-masses and station footprints co-located with observations. To complete a global 4D evaluation of ESM performance, the long-term stationary datasets should be complemented with surface or airborne transects, vertical profiles and satellite retrievals (e.g. van Noije et al., 2021).

With the competition between increasing spatial resolution and more detailed process descriptions in ESMs, data-driven approaches have been suggested to replace computationally intensive modules (Ahola et al., 2022). Whether through

470 emulation, neural networks or other machine learning techniques, the teaching and learning process requires comprehensive understanding of the model realm complemented with integrated observational suite (e.g. Schreck et al., 2022).

4.3 Integration of different approaches

475 **Figure 5: Combination of methods to integrate "bottom-up" and "top-down" insights on atmospheric aerosol and its interactions with clouds, as outlined within the FORCeS project (see forces-project.eu. Figure courtesy of Tinja Olenius.**

Combination of emerging long-term in-situ measurements, satellite data and process understanding bear a great potential for finding new ways to evaluate and constrain ESMs, and to reduce uncertainties in their projections (see Fig. 5, adopted from the FORCeS project). The inevitable spatial limitation related to in-situ observations can, to some degree, be overcome by

480 ensuring long enough temporal coverages and enhancing the number of representative data points (to be compared with satellites and models) this way (e.g. Isokääntä et al., 2022; Khadir et al., 2023).

Long-term, global in-situ observations are specifically useful in pin-pointing the model weaknesses and strengths, as well as in providing detailed observations with various techniques at well-defined altitudes, as opposed to sampling the entire atmospheric column. Long-term in-situ observations offer great opportunities to compare detailed process-level observations with satellite observations and large-scale models. Detailed measurements enable e.g. investigations of size-segregated trends in aerosol loadings in a regional context using both ESM and observational data (e.g. Leinonen et al., 2022). While the number of relevant data is steadily increasing, more long-term observations from under-sampled parts of the atmosphere (global south, highly remote areas) are needed. In addition, combining long-term in-situ measurements, satellite data, ESM model outputs and process understanding offers a great potential for finding new ways to evaluate and constrain the biosphere-climate feedbacks in ESMs, the magnitude of which is still highly variable between models (Thornhill et al., 2021). By separating the different processes (e.g. those that relate biogenic emissions and resulting aerosol concentrations to air temperatures and cloud properties), and by combining long-term observations with satellite data and multiscale modelling, one can facilitate the evaluation of the predictive abilities of the models (Blichner et al., to be submitted). This approach can help to isolate the impact of individual factors and improve our understanding of the underlying processes.

For example, the uncertainty in the effective radiative forcing due to aerosol-cloud interactions is governed by the cloud susceptibility to aerosol perturbations (Bellouin et al., 2020). This is split into two components which are (i) the response of the cloud droplet number concentration to aerosol perturbations – relevant for the radiative forcing due to aerosol-cloud interactions, also known as the Twomey effect – and (ii) the rapid adjustments in particular of cloud liquid water path and cloud fraction (Bellouin et al., 2020). In-situ long term aerosol and cloud observations enable investigations of cloud-susceptibility to aerosol perturbations. In-situ observations (both long-term and campaign-wise) and process understanding combined with ESM model outputs (or with satellites), facilitate pin-pointing specific processes or factors that should be improved in order to be able to describe the cloud activation and aerosol indirect forcing correctly in the models.

5 Future perspectives and possibilities

Currently, the speed of climate change along with its unpredictable consequences are challenging the capacities of existing observation systems. In addition, the ability to analyze the yet unknown questions and challenges, the "black swans" (Taleb, 2010), calls for comprehensive continuous observations. For example, COVID19 gave an unexpected opportunity to demonstrate the effect of exceptional reductions in anthropogenic emissions on air quality and climate (e.g. Gettelman et al., 2020; Wijnands et al., 2022). In this case the already running SMEAR-type, comprehensive measurements at the BUCT/AHL station in Beijing enabled us to investigate the atmospheric processes in detail. Other examples of this type of unusual and extraordinary events could be volcanic eruptions, gas pipe attacks, extreme weather events, forest fires, exceptionally dry periods, economic collapses or chemical weapons. All these events have both short- and long-term dynamic effects on air

quality and the climate system as well as on the functioning of societies. Also, the possibility of realized global tipping points, such as permafrost loss or boreal forest shift towards tundra, may lead to unexpected environmental episodes, events and feedbacks (Rockström et al., 2009, Kulmala et al., 2015, Lenton et al., 2019). The key questions are whether the majority of current observation systems contain sufficiently comprehensive set of variables to capture these events, and whether we have the preparedness to detect, analyze and quantify these events.

Recently proposed geoengineering approaches for mitigating global warming include a clear potential for large magnitude feedbacks which can have significant, yet unpredictable consequences to other processes. Risks related to uncontrolled geoengineering without international laws and the manipulation of the atmosphere highlight the value of continuous, comprehensive measurements detecting the changes. For example, operational Solar Radiation Modification (SRM) deployment would introduce new environmental and socio-economic threats like damaging the ozone layer and overcompensating climate change at regional scales (UNEP, 2023).

We need open big data to meet the present grand challenges, and we need to collect comprehensive data to be able to answer questions which do not exist today. The questions can be societal, economic or scientific, or any combination of these. To effectively collect, distribute and utilize big data, there are several key actions that needs to be considered:

Firstly, it is important to promote open data flows and storages globally via open access data platforms and structures. This can be achieved through optimizing data flows by also considering how to access and analyze the data at the storage site instead of transferring huge amounts of data. Advanced AI and data mining techniques should be employed to explore and utilize the data effectively. As importantly the in situ community should actively take part of the ongoing development of AI based Earth Virtualization Engines (EVE) which aims to animate the Earth observations, in situ and satellite data together, at 1 km scale for different users (Stevens et al. 2023). It is also important to transfer knowledge to make the data more accessible and to develop and provide examples and roadmaps for local data owners that highlights how to get merits via open data. The more local the data and data needs are, the more challenging it is to have them fully open. Therefore, it is crucial to demonstrate the benefits of offering open access to the data.

Secondly, ~~we need~~ global collaboration is needed to develop measurement protocols and data standards to reliably observe concentrations, fluxes and changes in the atmosphere and the environment. When using low-cost sensors, it is crucial to establish a proper calibration system that ensures data quality and traceability. Also, existing observation station types require calibrated sensors and enhanced harmonization. It is important to connect to existing harmonization actions by international organizations, such as the European Committee for Standardization (CEN), European environmental Research infrastructures and Network of Air Quality Reference Laboratories (AQUILA) etc. Several World Meteorological Organization working

groups are already active towards these goals. However, we need to go to the next level to make in-house processes more effective.

550   Finally, it is necessary to establish a hierarchy of stations ranging from cost-effective sensors (low cost) to comprehensive flagship stations, such as SMEAR – Stations to Measure Earth surface Atmosphere Relationships, by utilizing the knowledge and experience from the European Strategy Forum on Research Infrastructures (ESFRI) as well as from operational observation networks pertinent to different domains in the atmosphere – environment continuum. Within the next 20 years, we should have a station network utilizing the hierarchy of stations with three steps, namely Flagship stations, Median stations and low cost

555   sensors with enough Flagship stations scattered globally spatially and in ecosystem level to have enough representativeness of varying conditions. The comprehensive Flagship stations should be preferably part of GAW network with 500–1000 stations like SORPES, SIOS and SMEAR (Kulmala, 2018). The Median stations are high-end stations, but typically focusing on a specific topic (e.g. flux stations, AQ networks). Low cost sensors need calibration from Flagship and Median stations and utilizing of AI and 5G/6G/7G networks (Rebeiro-Hargrave et al., 2021).

560

Taking these actions will improve our understanding of the environment and our ability to respond to environmental challenges. An important question is who is willing to take the lead? Probably large international organizations are needed like WMO. In practice, we need to develop steps towards a GAW+ and maybe even to establish International climate /atmosphere institute. The institute should be multinational and multi-institutional research center following e.g. the model of CERN, but

565   focusing on the atmospheric and Earth system research.

This would be based on combining the experiences from WMO Global Atmospheric Watch (GAW) program (WMO 2017), COPERNICUS, and international *in situ* research infrastructures like the ESFRIs ICOS, ACTRIS and eLTER (see Section 1). The observation systems and research infrastructures, present standards, protocols and recommendations are consensus-based.

570   For example, essential atmospheric variables and data products management alone have been developed by several different actors, such as the Global Climate Observing System (GCOS) (WMO 2022) and GAW program (WMO 2017). These standardized systems have taken years to develop and are still in progress but need to be continued.

Under the WMO leadership, we should aim to the establishment of the global observatory for comprehensive data set(s) on

575   *weather, climate, water and environment.* This framework would provide a wide range of benefits, such as creating a real-world component and comparison for digital twin(s). It will also allow a proper WMO contribution to share integrated big datasets. The global observatory will provide a seamless connection between in-situ observations, remote sensing and multiscale model data. This will enable easy access and utilization of remote sensing products, such as inland water altimetry for rivers, lakes and reservoirs as well as arctic snow and ice cover. Furthermore, it would provide observational support for

580   global food forecasting through real-time dissemination of river level and discharge data, air quality forecasting and food and

water supply forecasting. It would also enable us to predict future climate and find out feedbacks and interactions between various environmental factors.

585 Once we have collected all these data, it is crucial that it is utilized effectively. To be able to use the big data, open access is typically needed. However, there are several barriers before the data can be used. The barriers include the lack of documentation, unknown data, misunderstood user needs, discipline specific jargon, bad and unusable interfaces, authorization problems, wrong terminology, training problems, unknown formats, difficulties in licensing and documentation, etc. To overcome the barriers of information, we need to have mutual trust and understanding of the needs, in addition to whcih we need to have access to the data to make new discoveries. This can be achieved by implementing the FAIR principles (Findable, 590 Accessible, Interoperable, Reusable) (Wilkinson et al., 2016), open data policies, proper knowledge transfer and conducting impact investigations e.g. IIASA.

It is worth noting that the most important reason to investigate multiple variables with continuous measurements is that we never know beforehand, when we will meet a 'black swan'. When we have comprehensive, continuous, open data, we can 595 analyze the data to study unexpected phenomena and answer to the upcoming challenges.

6 Conclusions

~~The need for comprehensive open data sets is obvious. Within the next 20 years, a new station network is needed, and the sooner the better.~~The need for comprehensive open data sets is obvious. The climate emergency and fast development of AI-based climate analysis force us to develop a new generation observation system. Within the next 20 years, we should have a 600 well-established station network, a "Global Earth Observatory", providing standardized data from different enviroments and scales.

Traditionally, and even in many cases today, there exist distinct infrastructures and their designated users, with experiences rather far from each other. Different research groups have typically their own instruments, raw data, data analysis methods and 605 publications. This is not an efficient way to meet grand challenges that can only be tackled with interdisciplinary approach. In the future, we need to utilize more joint efforts, including co-location of research infrastructures. Already to store raw data jointly, and to analyze it together in systematic ways, provides a big surplus. To have common data repositories for storing analyzed and published data is a big step forward.

610 One example on how comprehensive observations can be utilized is to solve air quality issues. In order to be able to understand the chemistry of air pollution, we need to observe multiple pollutants in existing air pollution cocktails (Kulmala, 2015). We should also remember that we spend 90% of our time indoors, and therefore also indoor air quality need to be understood. Air

quality is important e.g. for health effects, visibility and from the acidification point of view. The multiple pollutants include $PM_{2.5}$, size-resolved particle number, black carbon, O_3, NO_X, SO_2, CO, acids, various organic compounds etc., and their interactions and feedbacks. Our typical framework is a seamless chain from deep understanding to solutions, starting from observations and then continuing to understanding processes, feedbacks and interactions. These steps are needed to control air pollution and to improve air quality.

In basic and applied research, we make new discoveries and new knowledge from the resources we have with the money we have at our exposal. New discoveries and knowledge often lead to innovations and with innovations people make money from that knowledge. Innovations and new knowledge are a key for society to maintain and enhance wellbeing and this way the circle closes.

It is crucial to utilize multidimensional, multidisciplinary, multiscale approach to be able to answer questions related to grand challenges. It is also important to have a clear and ambitious vision from deep understanding to practical solutions. Also, we need seamless chain to connect measurements, modelling and theory as well as from research to innovations, economic growth and human wellbeing.

The main benefits that research community can gain from using integrated research infrastructure approach includes higher-quality science and higher visibility and recognition from the society and its various stakeholders and higher amount of scientific users both nationally and internationally. Moreover, collaboration possibilities will be enhanced, including feedbacks between domains, landscape analysis, up- and downscaling. Detailed experiments and observations can support each other, so that new observational and numerical methods can be developed. The improved utilization of data flows and synergies in data use are foreseen.

In order to meet grand challenges and answer open scientific, societal and economic questions, we need to build upon a network of domain-specific research infrastructures, such as ACTRIS, eLTER and ICOS. We need to acknowledge that for example ACTRIS is already integrating several subfields, namely aerosol in-situ, trace gas and cloud in-situ observations and ground-based remote sensing of aerosols, clouds and trace gases. The human-environment relationship, i.e., the social-ecology research, is part of the transdisciplinary approach in eLTER, enabling policy-relevant research and interaction with society at different scales. The SMEAR concept in essence includes co-location and integration of the observations performed in the domain specific environmental RIs. A further connection to and integration with e.g., health and societal data are needed. Furthermore, we need excellent science, with high quality, critical mass and interdisciplinary research as well as education and training, i.e. knowledge exchange. We need to contribute to innovation ecosystem and have continuous, long-term dialogue with policy makers. Internationally, this enables clear contributions to science diplomacy based on integrated scientific viewpoint.

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