Broadening Impact of Field Campaigns

Integrating Meteorological and Chemical Observations

Gretchen L. Mullendore, Mary C. Barth, Petra M. Klein, and James H. Crawford

ABSTRACT: Historically, atmospheric field campaigns typically focused on either meteorology or chemistry with very limited complementary observations from the other discipline. In contrast, a growing number of researchers are working across subdisciplines to include meteorological and chemical measurements when planning field campaigns to increase the value of the collected datasets for subsequent analyses. Including select trace gas measurements should be intrinsic to certain dynamics campaigns, as they can add insights into dynamical processes. This paper highlights the mutual benefits of joint dynamics–chemistry campaigns by reporting on a small sample of examples across a broad range of meteorological scales to demonstrate the value of this strategy, with focus on the Deep Convective Clouds and Chemistry (DC3) campaign as a recent example. General recommendations are presented as well as specific recommendations of chemical species appropriate for a range of meteorological temporal and spatial scales.

<https://doi.org/10.1175/BAMS-D-19-0216.1>

Corresponding author: Gretchen Mullendore, gretchen@ucar.edu

Supplemental material: <https://doi.org/10.1175/BAMS-D-19-0216.2>

In final form 15 September 2020

©2021 American Meteorological Society

For information regarding reuse of this content and general copyright information, consult the [AMS Copyright Policy](#).

Significant economic and personnel resources are invested in atmospheric sciences field campaigns each year. This investment is crucial for the furthering of our scientific understanding of how the atmosphere works and ultimately helps us improve our ability to forecast hazards associated with weather, climate, and air quality. Many campaigns target hypotheses that emphasize either atmospheric dynamics or atmospheric chemistry, but testing these hypotheses relies on a balance of both meteorological and chemical observations. Often, this balance can be improved by adding relatively low-cost chemical and/or meteorological measurements to the primary proposed suite of observations. This calls for careful consideration in the initial planning of field campaigns that considers complementary chemical measurements for dynamical campaigns and complementary meteorological measurements for chemistry campaigns. Such consideration should also be extended to allow broader involvement from these two research communities which ultimately allows more and better science to be done. Leveraging the investment already made in field campaigns to include a few additional measurements, e.g., measurements of carbon monoxide (CO) and ozone (O₃) to distinguish tropospheric and stratospheric air or a wind profiler to provide insight on PBL dynamics, results in an increase in scientific value that far outweighs the extra investment. This approach was part of the planning for the Deep Convective Clouds and Chemistry (DC3; Barth et al. 2015) campaign, which will be described in more detail in the third section and can be used as an example case for future campaigns.

Modeling studies have employed passive tracers to understand flow fields for decades. However, validating these model findings with observations, particularly in clear-air conditions, remains a challenge. Whether complex flow in terrain, fluxes across boundaries (e.g., planetary boundary layer entrainment, troposphere–stratosphere exchange), storm motions, or inertia–gravity waves, researchers rarely have enough observations to characterize the full flow field (including turbulent mixing). Several recent national workshops/reports have highlighted the limitations of traditional observational strategies for furthering our understanding of the dynamical flow and mixing (e.g., Geerts et al. 2017, 2018; National Academy of Sciences 2017); these same reports highlight the value of tracers in constraining dynamics. Chemical tracers can provide insight into “unseen” motions, unveil the vertical structure of the planetary boundary layer (PBL), and identify the source regions of different air masses.

The National Academies of Sciences (2016) report on advancing atmospheric chemistry highlights the need to understand the coupling between dynamics and chemistry. The addition of atmospheric state variables to chemistry-focused field campaigns provides information on the dynamical state to quantify the uncertainty introduced by meteorological phenomena (i.e., uncertainty in chemical processes are more valuable if meteorology uncertainty is included). Further, atmospheric chemistry studies could show exaggerated variance in results if not properly composited by the meteorological regime.

More deliberate integration of meteorological and chemical observations will lead to improved collaborations between the dynamics and chemistry communities. While there are many examples of valuable research resulting from strong collaboration between dynamists and chemists, these fields can still benefit from closer interaction. A fundamental level of understanding of both fields is valuable to all atmospheric scientists, from students to senior-level researchers. Increased collaboration is key to helping us further understanding

in both fields, e.g., the role that wildfire smoke has on temperature forecasts or the role that precipitation has on air quality.

The goal of this article is to bring awareness to how the integration of meteorological and chemical perspectives benefits field campaigns. The following discussions provide a high-level background on the relevant connections between meteorology and chemistry as well as historical examples where joint investigation of meteorology and chemistry have been accomplished to good effect. Our target audience is not the group of researchers already doing this type of work, but the meteorologists and chemists who rarely think about using observations outside of their own field. As such, the tracer information provided here is not a comprehensive listing of all valuable tracers, but instead offers examples of chemical tracers that have already been successfully used to gain insight into atmospheric flows.

Spatial/temporal scales of meteorological regimes and chemical tracers

Figure 1 shows atmospheric spatial and temporal scales of various trace gases alongside meteorological phenomena. While not exhaustive, this figure gives guidance on common chemical measurements that can be useful for understanding complex flow fields of a given scale. Having a chemical lifetime longer than the temporal scale of the meteorological phenomenon (i.e., effectively passive) allows researchers to ignore effects of chemical transformations and instead attribute changes in concentration to mixing along the air parcel trajectory. The environmental origin of a constituent along with lifetime can also dictate the value of a measurement. For instance, isoprene and monoterpenes are associated mainly with emissions from vegetation under heat stress, while dimethyl sulfide (DMS) and CH_3I are emitted mainly in marine environments. All of these are short lived enough that their presence in the free troposphere can only occur through rapid vertical transport making them excellent indicators of air from recent convection. The other hydrocarbon species in the figure are largely associated with human activity through fossil fuel exploration and use. Given their relative lifetimes, their changing ratios can provide a sort of chemical clock for understanding the balance between chemistry and mixing as air is transported from source regions. The isotopic constituents, Be-7 and Pb-210, have specific source regions with Be-7 originating from cosmic ray activity in the stratosphere and Pb-210 from the rapid decay of terrestrial emission of Rn-222. While they have long half-lives, they are also subject to wet removal, thus their distributions provide valuable

Fig. 1. Time and spatial scales of various trace gases and meteorological phenomena. Time scales for O_3 and SO_2 are from global model estimates, CH_3I are based on photodecomposition rate, and Rn-222 and Be-7 on radioactive decay. Time scales of all other trace gases are based on reactivity with OH, where OH is assumed to be 2×10^6 molecules cm^{-3} and temperature is 298 K. Spatial scales of trace gases are based on a 2 m s^{-1} wind. C_3H_6 = propene; CH_3CHO = acetaldehyde; C_2H_4 = ethene; DMS = dimethyl sulfide; C_5H_{12} = pentane; SO_2 = sulfur dioxide; C_4H_{10} = butane; CH_3I = methyl iodide; C_3H_8 = propane; C_2H_2 = acetylene; O_3 = ozone; C_2H_6 = ethane; CO = carbon monoxide; Be-7 = beryllium-7; and Pb-210 = lead-210. Compounds with the same symbol indicate the possibility that they could be measured with a single instrument. Specific measurement techniques for detecting each compound are provided in the supplemental material. STE = stratosphere–troposphere exchange; PBL = planetary boundary layer.

constraints on transport between the stratosphere and troposphere in the case of Be-7 and large-scale circulations from terrestrial to marine environments for Pb-210. Current measurement capabilities enable measurement of the constituents in Fig. 1 across their full range of atmospheric variability down to background levels, and many can be measured at one second time resolution (e.g., O₃, CO, SO₂, C₂H₆, isoprene, toluene), but this would require multiple instruments. To measure the full suite of hydrocarbons simultaneously, whole air sampling (WAS) remains the most effective method along with online gas chromatography mass spectrometry (GC-MS) measurements that can provide in-flight data.

The online supplementary materials (<https://doi.org/10.1175/BAMS-D-19-0216.2>) include a table quantifying the temporal and spatial scales for these trace gases as well as instrument techniques used to measure them and useful references. There are many avenues to learn more about instruments that might be of use to a particular campaign. We recommend contacting colleagues at universities, at the national laboratories (NASA, NOAA, DOE), or other research institutes (e.g., NCAR, MPI-Chemie). Additionally, program and facility managers at funding agencies can help connect researchers to instrument teams.

The examples given here are organized into frequently studied phenomenological categories that span a range of scales: PBL flows, moist convection, and tropical cyclones, monsoons, and long-range transport. Each section below gives specific examples of research projects for each scale category that have made use of tracer chemicals from field campaigns. We focus this paper on gas species, but there are many examples of aerosols also serving as effective tracers, such as air masses impacted by fires or volcanic eruptions. We also do not include water isotopes in the current article, but they may also be useful as water isotopes become more feasible for airborne research. Water isotopes have been used extensively to identify processes such as entrainment and exchanges with the ocean/land (e.g., Bailey et al. 2015, 2019).

Planetary boundary layer flows. Air motions in the PBL span spatial (L) and temporal (T) scales ranging from fluxes in the surface layer and flow in canopy layers ($L \approx 1\text{--}200$ m, $T \approx 1$ s–30 m) to the diurnal cycle of PBL growth and decay ($L \approx 200$ m–3 km, $T \approx 30$ min–24 h). Air quality studies in the boundary layer require detailed observations of meteorology, emissions, and chemistry (e.g., Crawford and Pickering 2014) to understand dispersion and deposition of pollutants (see “Chemistry campaigns” section for more discussion). In studies of PBL dynamics, tracers can be used to characterize turbulence and entrainment processes.

Many studies have used ozone as the conserved tracer to study PBL entrainment over the oceans (e.g., Lenschow et al. 1988; Paluch and Lenschow 1991). The steep increase in ozone concentration above the PBL allows calculation of entrainment and characterization of turbulence regimes. For example, Paluch and Lenschow (1991) were able to use ozone measurements at cloud base to distinguish between PBL downdrafts associated with cloud-top radiative cooling (low ozone) and those caused by mixing with the free troposphere (high ozone). Studies such as Lenschow et al. (1988) have also used back trajectories to assess source regions of the sampled areas. Back trajectories demonstrate a valuable coupling of dynamical and chemical measurements. While there is some uncertainty in both the wind fields used to calculate the air parcel trajectories and the assumptions made to calculate the evolution of chemical conditions in those parcels, checking one method against the other improves confidence in parcel pathways.

Over land, ozone is also widely used but not assumed conserved. Near-surface ozone concentrations, ideally supplemented by vertical ozone profiles, can provide insights about turbulence, stability and long-range transport in the nocturnal PBL (Banta et al. 1998; Zhang and Rao 1999; Klein et al. 2014). Further, ozone is often combined with another tracer, such as CO₂, to conduct tracer–tracer correlations that can provide further detail on

the evolution of boundary layer structure and mixing events (e.g., Berkes et al. 2016). For these studies to be successful, expertise on both meteorology and chemical reactions/fluxes are needed to properly constrain the observations. For example, when ozone could not be assumed steady state (e.g., Patton et al. 2011; Klein et al. 2014), several additional chemical (NO , NO_x) and meteorological measurements were included to understand the impacts of advection, vertical mixing, and chemistry on ozone flux (Fig. 2). Figure 2a shows a time period with lower wind speeds (contours) and lower friction velocities (black dots) during the nocturnal hours than the case shown in Fig. 2b. The higher stability and reduced mixing in the decoupled surface layer of the first case result in more ozone depletion via NO titration reactions (NO_x ; dashed line) and hence markedly lower nocturnal surface ozone concentrations (solid line). During the second time period (Fig. 2b) a strong low-level jet develops right after sunset and friction velocities remain high throughout the night. Ozone is actively mixed down to the ground, slowing the nighttime depletion of ozone at the surface. Complementary Weather Research and Forecasting Model with Chemistry (WRF-Chem) simulations provided further evidence that during the first night ozone concentrations were high in the residual layer that was decoupled from the very depleted stable surface layer while during the second night the ozone depletion that did occur was throughout a much deeper layer.

Moist convection. Moist convection spans spatial and temporal scales from single cells to large organized systems such as mesoscale convective complexes ($L \approx 1\text{--}500$ km, $T \approx 30$ min–24 h). Tracers have been used to characterize the amount of mass transport from the boundary layer in deep convection, identify the age of air in convective outflows, and analyze convectively forced mixing across the tropopause. Because PBL air usually has a much higher CO concentration than air in the upper troposphere and lower stratosphere (UTLS), CO plumes in the UTLS have long been used to identify air influenced by convection (e.g., Dickerson et al. 1987). If the concentrations of CO at cloud base are well constrained, CO concentrations aloft can also provide insight into the amount of entrainment occurring in the updraft (i.e., undiluted concentration indicates negligible entrainment). Correlating CO concentrations (proxy for PBL air) and ozone concentrations (proxy for stratospheric air) is frequently used as a method to quantify convective mixing of tropospheric and stratospheric air (e.g., Huntreiser et al. 2016). For example, Fig. 3 shows similar values of inflow CO (O_3) to midanvil outflow CO (O_3), indicating convective transport; O_3 mixing ratios > 120 ppbv just on the north side of the anvil coincident with low CO shows stratospheric air at altitudes of the anvil. Chemical tracers can also elucidate previously undocumented transport pathways, such as wrapping of stratospheric air around the edges of mesoscale convective systems (MCSs; Pan et al. 2014).

Fig. 2. The O_3 (solid lines) and NO_x (dashed lines) concentrations at the Oklahoma City monitoring site, during (a) 17–18 and (b) 25–26 Jul 2003, together with wind profile data (color map) collected as part of the Joint Urban 2003 (JU2003) tracer experiment. The black dots show friction velocities observed at 37 m AGL on a tower (to fit the axis scale, cm s^{-1} was chosen as the unit for friction velocities in these plots). (Adapted from Klein et al. 2014, their Fig. 6.)

Fig. 3. Maximum column radar reflectivity (dBZ) and (a) CO (ppbv) and (b) O₃ (ppbv) mapped for the region of the 22 Jun 2012 DC3 severe convection. The colored circles indicate the mixing ratios of the trace gas in the inflow region (marked) and outflow region of the storms. Radar reflectivity is for 2312 UTC 22 Jun, at the time of the inflow measurements. Arrows show the wind direction measured on the two aircraft (NSF/NCAR GV and NASA DC-8). Also marked is a smoke plume from the High Park wildfire.

While chemical tracers are extremely valuable for studying air motions, one needs to be aware of possible contamination of the tracer signal and strategies to overcome these challenges. For example, plumes of CO can exist downwind of convective outflow that are actually from long-range transport of pollutant and/or biomass burning and not from the convection being studied. In Fig. 3, higher outflow CO than inflow CO indicates that a smoke plume was ingested by the storm and transported into the anvil. In addition to contamination issues, there can be a significant amount of variability of CO in the PBL, including regions with very clean conditions, i.e., very weak CO signals. Significant variability in background state can happen for a wide range of tracer species. This challenge, which occurs across all scales of motion, can be mitigated with additional tracer species included in the observational suite and/or carefully designed modeling studies.

With a more extensive suite of chemical measurements, “age of air” studies can be conducted to investigate multiple transport pathways (e.g., Waugh and Hall 2002). Luo et al. (2018) used 42 volatile organic compounds (VOCs) encompassing a wide range of lifetimes to estimate tropical convective transport times to the upper troposphere (see their Table 1 for a list of compounds). This study was able to show that the dominant mode of transport of VOCs from the marine boundary layer to the upper troposphere was convective mass flux.

Tropical cyclones, monsoons, and long-range transport pathways. Tropical cyclone and monsoon circulations span spatial and temporal scales from the deep convection and biomass burning events embedded within these features to the mean state impact of these systems on the global circulation ($L \approx 100\text{--}1,000$ km, $T \approx 12$ h–several months). While the mean state circulation and chemical signatures of these phenomena (and of long-range transport) can be assessed using remote sensing techniques such as satellite, in situ campaigns are needed to understand the perturbation events that lead to the mean state. These perturbation events may be identified by field campaigns designed to observe short-term events and/or small-scale features. For example, Gottschaldt et al. (2017) used tracer-tracer relations of CO, HCl, and O₃ to understand the relative contributions of deep convection and mixing with stratospheric air to the Asian summer monsoon anticyclone (Müller et al. 2016).

Other examples of small-scale events embedded in large-scale flow would be intercontinental pollution transport and vertical transport of tracers in the tropical UTLS. Cooper et al. (2004) analyzed aircraft measurements of a suite of chemicals to study vertical mixing by a warm conveyor belt (WCB) and subsequent long-range transport from Asia to North America. Using a combination of trajectory and chemical analyses, three different general transport pathways were identified for the measurements, including the previously unobserved mechanism of pollutants being processed through multiple WCBs.

Konopka et al. (2007) and Sargent et al. (2014) both used models combined with in situ chemical measurements to investigate the processes impacting transport and mixing of air into the tropical tropopause layer, highlighting the significant contribution from deep convection. Pommrich et al. (2014) combined both satellite and in situ measurements to assess simulated vertical transport to the tropical UTLS. Overall, the model captured the large-scale vertical transport, but the model significantly underpredicted enhanced CO in convective outflow, a feature only sampled by in situ observations.

Meteorological and chemical measurements have also enhanced studies of tropical cyclones. Newell et al. (1996) used multiple tracers including DMS (marine boundary layer proxy) and O₃ (stratosphere proxy) to better understand airflow in Supertyphoon Mireille (Hoell et al. 1996). Additionally, tracer ratios such as acetylene (C₂H₂) to CO were used to determine the age of the inflow air. McKeen et al. (1996) used hydrocarbon ratios from the same campaign to understand the relative effects of air mass mixing and photochemistry.

Field campaigns and atmospheric models. Although this article is focused on observational field campaigns, we want to briefly highlight the important role of atmospheric models in observational work. In fact, the majority of the field campaigns described in this article had a strong modeling component. Often, field campaigns are motivated by deficiencies in weather, climate, or atmospheric chemistry models. As discussed in the introduction, observational studies can help us investigate and refine model processes that are unresolved, i.e., motions and mechanisms that must be estimated at the scale of the model. Tracer measurements are an exceptional addition to closing budgets on unresolved motions. For example, a researcher may want to run back trajectories to understand the evolution of a particular flow field. Modeled winds can be used to calculate the parcel pathways, but some portion of the mixing processes are always unresolved. Tracer concentrations along that pathway, however, will change based on subgrid-scale mixing (and other unresolved processes). By simulating tracers, we can compare observed concentrations to the modeled concentrations and quantify how well the parameterized processes are captured in the model.

Further, simulated tracers can help in designing observational strategies. For example, regions of the simulated flow field where parcel concentrations rapidly change along a streamline indicate where small-scale processes (e.g., chemical reactions, dynamical mixing, microphysical scavenging) are particularly active. This information can motivate targeted observations to more finely instrument those active areas and/or careful sampling of inflow and outflow regions for better constrained budget calculations.

Finally, postcampaign, models can be used to understand the physical and dynamical processes for tracer evolution and transport. With close collaboration between meteorological and chemical experts, an atmospheric model with appropriate grid spacing can then be used to systematically investigate and identify the dominant processes responsible for the observed behavior (e.g., Klein et al. 2014; Phoenix et al. 2020).

For example, meteorological and chemical observations collected during the Joint Urban 2003 (JU2003) campaign (Allwine 2004), in the Oklahoma City region provided insight about the impact of low level jets (LLJs) on PBL mixing processes (Fig. 2). The O₃ and NO_x observations suggested several important transport and mixing regimes that affected the diurnal cycle of

near-surface O_3 concentrations, yet the observations could not fully constrain the problem. By using simulations to understand the important dynamical (long-range transport and vertical stability/mixing) and chemical processes (photochemistry and titration reactions), Klein et al. (2014) could explain why the markedly different observations occurred during the different time periods. Further, uncertainties in this modeling study clearly showed what would be needed in future campaigns.

Chemistry campaigns. The focus in the above sections has been on the benefit of chemical tracers to our dynamical understanding. Likewise, interpretation of detailed chemical measurements can derive great benefit from small investments in meteorological measurements. For example, detailed chemical understanding requires simultaneous measurements of many compounds to inform fingerprinting of emission sources and the complex chemical processing that leads to the formation of secondary pollutants and oxidation products. Such comprehensive measurements can only be accomplished in situ and thus offer information that is limited to a time series at a specific point on the ground or along a flight path through the atmosphere. With such a limited perspective, these chemical observations are vulnerable to misinterpretation in the absence of information regarding the conditions aloft at a ground site or above and below a research aircraft. While in situ meteorology is routinely collected (e.g., temperature T , pressure P , relative humidity, winds) at ground sites and on research aircraft, investment in multidimensional information through soundings (sondes, tether sondes, drop sondes) or remote sensing (lidars, ceilometers, sodars, wind profilers, etc.) can provide invaluable context on the same phenomena discussed above, but with their relevance to detailed chemical processes. For instance, recent campaigns focused on air quality [Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) and Korea–U.S. Air Quality (KORUS-AQ)] employed ceilometers, micropulse lidars, and tether sondes at ground sites to monitor diurnal changes in PBL mixing, sea breeze fronts, and other phenomena affecting chemical gradients between the surface and lower atmosphere. Without this information, many changes in surface level quantities could not be fully understood in terms of the competition between emission fluxes and their ventilation aloft. A good example of this can be found during the DISCOVER-AQ sampling in the Denver area showing that vertically integrated abundances of nitrogen dioxide in the PBL can increase by a factor of 2 in the morning while surface values decrease by the same amount as traffic emissions are mixed into a rapidly deepening PBL (Crawford et al. 2016). Such rapidly changing conditions also lead to substantial vertical gradients within the PBL for short-lived species with variable emissions. This information is of crucial importance to understanding ozone production and the rates of chemistry not only at the surface, but also aloft. Variable ozone production efficiencies at different altitudes determine the integrated ozone production in the PBL that ultimately affects surface ozone (Zhang et al. 2016).

Recipe to implement: Lessons learned from DC3

The DC3 field campaign provides an example of the successful integration of both meteorology and atmospheric chemistry measurements (Barth et al. 2015). Here, we describe the activities associated with DC3 that made it successful as well as some lessons learned.

The DC3 field campaign aimed to understand how thunderstorms affect the chemical composition of the troposphere and lower stratosphere. While atmospheric chemistry transformations were analyzed, the key processes examined were physical and dynamical ones that affected trace gases and aerosols. These processes included transport, scavenging of trace gases and aerosols by precipitation, lightning-generated nitrogen oxides, and mixing

between the stratosphere and troposphere. Thus, it was necessary to bring together three atmospheric science subcommunities: atmospheric chemistry, atmospheric electricity, and storm dynamics and physics, including experts in radars.

The DC3 field experiment was designed by four principal investigators with extensive input given by a science steering committee and, during precampaign workshops, by research scientists from the three atmospheric science subcommunities. The expertise of the four principal investigators (PIs) ranged from radar and mesoscale meteorology, precipitation physics, and atmospheric electricity to interactions between clouds and chemistry and atmospheric photochemistry. Bringing together PIs from the primary focus areas of DC3 science was important for generating a valuable measurement suite.

Day 1 and day 2 execution. To address convective processing of trace gases, the DC3 project sampled the inflow and outflow regions of thunderstorms in the observation area of research-grade, ground-based radars and lightning mapping arrays, which documented storm structure and kinematics, and lightning activity, respectively. Determining the location for the daily observations was based on meteorological forecasts performed by the DC3 forecast team. The DC3 aircraft that conducted coordinated sampling reached the region of interest usually before storms developed so that they could characterize the composition of the troposphere unaffected by storm activity. Where to position the aircraft during storm activity was determined by meteorology (winds primarily), but analysis of trace gases observations was used to identify specific characteristics of the air mass (e.g., influence of anthropogenic or biomass burning plumes).

Key trace gas measurements on the aircraft included CO, O₃, NO, NO₂, alkanes, and alkenes. Using ratios of trace gases with similar photochemical lifetimes (e.g., i-C₄H₁₀ and n-C₄H₁₀, or i-C₅H₁₂ and n-C₅H₁₂) provides a way to connect air masses from the PBL to the upper troposphere. These ratios are unique depending on the emission sources in each region (e.g., urban versus oil and gas production). If the ratio in the upper troposphere is similar to the ratio in the PBL, then that is strong evidence that the air masses are connected via convection and/or advection processes. Using ratios of trace gases with different photochemical lifetimes provides a way to estimate the photochemical age of the air mass. Trace gases that are not soluble, have a long chemical lifetime compared to the transport time, and have a well-defined source such as the PBL (e.g., CO, alkanes, alkenes) can be used to estimate entrainment in convective clouds (Fried et al. 2016; Barth et al. 2016).

Challenges associated with these measurements for addressing convection are twofold. As discussed above, CO is often used to identify PBL air and to estimate entrainment in clouds. However, its background mixing ratios in the free troposphere have increased such that there is not a strong difference between clear air CO and in-cloud CO. Thus, alkanes and alkenes can instead be used for making these calculations. The VOC measurements are not fast, taking 10 s [for proton transfer reaction spectrometry (PTRMS)] and up to a couple minutes (for collecting air that is analyzed by gas chromatography in the laboratory), making it challenging to collect reliable measurements in small clouds (e.g., airmass thunderstorms where aircraft transect times across the top of the storm lasts ≈10 s). Having measurements of trace gases with a spectrum of time scales and measurement frequency increases the ability to achieve successful science analysis of the observations.

The DC3 project included the goal of obtaining measurements in convective outflow after it had chemically aged for several hours (i.e., on day 2). To assess the feasibility of finding convective outflow air in a region both easily reached by the aircraft and also safe to fly, the DC3 forecast team analyzed forecasts of artificial boundary layer and lightning NO_x tracers on the morning of day 1 to learn where the forecasted convective outflow was expected to be on day 2. However, those outflow forecasts were often highly uncertain due to the poor

predictability of convection and its outflow direction. Therefore, during and after the day-1 convection was sampled, trajectory modeling initialized from the observed convection was used to determine more accurately the location of the day-2 convective outflow. The aircraft flew to the forecasted region of convective outflow, using trace gases to help identify the type of air mass being sampled. This approach worked well for only one of the five day-2 sampling events. The other events were not as successful because of changing meteorology between forecasts and measurement deployment.

The value of an integrated field campaign: Example case. The 21 June 2012 case from DC3 exemplified the benefit of including meteorologists and atmospheric chemists in the experimental design and execution of the field experiment. During the field experiment, the PIs and lead forecaster realized that a possible way to sample the chemical aging of a convective outflow air mass would be by taking measurements in the outflow of an MCS and to remain in that air mass as it decayed during the day. This approach was not part of the original experimental design and brought together the best of the meteorology and chemistry because an MCS would still be active early in the morning when sampling would begin such that the convective outflow was easily located, and the aircraft would probe the air mass during the daytime when photochemistry is most active. Thus, two DC3 aircraft were deployed, one beginning its flight before 0700 local time and the second beginning its flight 4–5 h later, totaling ≈ 10 h in the convective outflow plume. Remaining in the air mass was achieved by using the aircraft winds and potential temperature observations. Although not used, observations of trace gases that are long lived could also be utilized to confirm that the aircraft remained in the air mass.

This initial attempt to sample the effects of an MCS on chemical composition of the upper troposphere worked well. Many of the measured trace gas mixing ratios changed as previous modeling studies predicted (Brune et al. 2018). The experience of conducting this quasi-Lagrangian approach was positive and the approach is recommended for future field experiments studying convective outflow plumes.

Summary and recommendations

This article has presented a case for more widespread use of tracer measurements in meteorological campaigns. Figure 1 provides recommendations for useful chemical tracers. In planning a campaign, one should consider the temporal scale of the flow field of interest. Chemical species can act as passive tracers if their lifetimes are much longer than the temporal scale of interest (and there is a reliable and predictable source of that tracer and reasonable expectations for negligible contamination from additional sources). Additionally, species with a range of temporal lifetimes can be used for age of air studies. We also advocate for more widespread and consistent inclusion of basic meteorological measurements in atmospheric chemistry campaigns. For any atmospheric chemistry campaign, we recommend measurements of temperature, wind speed and direction, pressure, and humidity with emphasis on local, regional, and vertical sampling, particularly in the PBL (see “Chemistry campaigns” section). These measurements should be taken at temporal and spatial scales appropriate for the atmospheric processes impacting the chemistry being studied (and not already resolved by standard observations or reanalysis data). Partnering with dynamicists is important for identifying these impactful processes and scales; partnering with meteorological instrument experts is important for designing observational strategies that will be effective even in challenging observational conditions (e.g., cloudy conditions can reduce effectiveness for many instruments).

As discussed in the introduction, we recognize that a growing number of scientists are already combining expertise across meteorology and chemistry at both the campaign stage and with subsequent analyses. We see this trend continuing in many upcoming campaigns

[e.g., Asian Summer Monsoon Chemical and Climate Impact Project (ACCLIP), www2.aoml.ucar.edu/acclip; Dynamics and Chemistry of the Summer Stratosphere (DCOTSS), dcotss.org]. Additionally, basic chemical measurements and analysis are being used in meteorological campaigns and enhanced meteorological measurements being used in chemistry campaigns [e.g., Cloud System Evolution in the Trades (CSET); Albrecht et al. 2019; Bretherton et al. 2019]. This article is not a comprehensive review of these many prior and future studies. Our goal instead was to use selected examples to highlight and briefly explain the approach for those who are not considering the full suite of observational options.

In summary, we advocate more collaboration between the meteorological and chemistry communities, to the benefit of both. Of course, adding any additional observational platforms to a field campaign increases the costs of the campaign, but with judicious choices the costs can provide substantial benefits to the science analyses. With active collaboration between scientists of varied expertise (as in DC3), we can design campaigns that provide increased value to the entire scientific community.

Acknowledgments. This paper was inspired by the outcomes of the NSF/NCAR C-RITE workshop (www.eol.ucar.edu/c-rite-workshop). We thank Bart Geerts and two anonymous reviewers for their valuable comments. We thank Xiaoming Hu for updates to Fig. 2 and the instrument PIs for the data shown in Fig. 3, including Tom Ryerson (O_3), Glen Diskin (CO), and for the composite radar data provided by Cameron Homeyer. The National Center for Atmospheric Research is supported by the National Science Foundation.

Data availability statement. All analyses and figures based on previously published results. Data from the DC3 campaign can be accessed at <http://catalog.eol.ucar.edu/dc3/>.

References

- Albrecht, B., and Coauthors, 2019: Cloud System Evolution in the Trades (CSET): Following the evolution of boundary layer cloud systems with the NSF–NCAR GV. *Bull. Amer. Meteor. Soc.*, **100**, 93–121, <https://doi.org/10.1175/BAMS-D-17-0180.1>.
- Allwine, K., 2004: Overview of Joint Urban 2003—An atmospheric dispersion study in Oklahoma City. *Symp. on Planning, Nowcasting, and Forecasting in the Urban Zone*, Seattle, WA, Amer. Meteor. Soc., J7.1, https://ams.confex.com/ams/84Annual/techprogram/paper_74349.htm.
- Bailey, A., D. Noone, M. Berkelhammer, H. C. Steen-Larsen, and P. Sato, 2015: The stability and calibration of water vapor isotope ratio measurements during long-term deployments. *Atmos. Meas. Tech.*, **8**, 4521–4538, <https://doi.org/10.5194/amt-8-4521-2015>.
- , H. K. A. Singh, and J. Nusbaumer, 2019: Evaluating a moist isentropic framework for poleward moisture transport: Implications for water isotopes over Antarctica. *Geophys. Res. Lett.*, **46**, 7819–7827, <https://doi.org/10.1029/2019GL082965>.
- Banta, R. M., and Coauthors, 1998: Daytime buildup and nighttime transport of urban ozone in the boundary layer during a stagnation episode. *J. Geophys. Res.*, **103**, 22 519–22 544, <https://doi.org/10.1029/98JD01020>.
- Barth, M. C., and Coauthors, 2015: The Deep Convective Clouds and Chemistry (DC3) field campaign. *Bull. Amer. Meteor. Soc.*, **96**, 1281–1309, <https://doi.org/10.1175/BAMS-D-13-00290.1>.
- , and Coauthors, 2016: Convective transport and scavenging of peroxides by thunderstorms observed over the central U.S. during DC3. *J. Geophys. Res. Atmos.*, **121**, 4272–4295, <https://doi.org/10.1002/2015JD024570>.
- Berkes, F., P. Hoor, H. Bozem, D. Kunkel, M. Sprenger, and S. Henne, 2016: Airborne observation of mixing across the entrainment zone during PARADE 2011. *Atmos. Chem. Phys.*, **16**, 6011–6025, <https://doi.org/10.5194/acp-16-6011-2016>.
- Bretherton, C. S., and Coauthors, 2019: Cloud, aerosol, and boundary layer structure across the northeast Pacific stratocumulus–cumulus transition as observed during CSET. *Mon. Wea. Rev.*, **147**, 2083–2103, <https://doi.org/10.1175/MWR-D-18-0281.1>.
- Brune, W. H., and Coauthors, 2018: Atmospheric oxidation in the presence of clouds during the Deep Convective Clouds and Chemistry (DC3) study. *Atmos. Chem. Phys.*, **18**, 14 493–14 510, <https://doi.org/10.5194/acp-18-14493-2018>.
- Cooper, O. R., and Coauthors, 2004: A case study of transpacific warm conveyor belt transport: Influence of merging airstreams on trace gas import to North America. *J. Geophys. Res.*, **109**, D23S08, <https://doi.org/10.1029/2003JD003624>.
- Crawford, J., and K. Pickering, 2014: DISCOVER-AQ: Advancing strategies for air quality observations in the next decade. *EM Magazine*, Vol. 64, No. 9, 4–7.
- , and Coauthors, 2016: Multi-perspective observations of nitrogen dioxide over the Denver area during DISCOVER-AQ: Insights for future monitoring. *EM Magazine*, Vol. 66, No. 8, <https://pubs.awma.org/flip/EM-Aug-2016/crawford.pdf>.
- Dickerson, R. R., and Coauthors, 1987: Thunderstorms: An important mechanism in the transport of air pollutants. *Science*, **235**, 460–465, <https://doi.org/10.1126/science.235.4787.460>.
- Fried, A., and Coauthors, 2016: Convective transport of formaldehyde to the upper troposphere and lower stratosphere and associated scavenging in thunderstorms over the central United States during the 2012 DC3 study. *J. Geophys. Res. Atmos.*, **121**, 7430–7460, <https://doi.org/10.1002/2015JD024477>.
- Geerts, B., M. Barth, A. Detwiler, P. Klein, W.-C. Lee, P. Markowski, G. Mullendore, and D. Raymond, 2017: Requirements for In Situ and Remote Sensing Capabilities in Convective and Turbulent Environments (C-RITE) community workshop. Earth Observing Laboratory Final Rep., 46 pp., <https://doi.org/10.5065/D6DB80KR>.
- , and Coauthors, 2018: Recommendations for in situ and remote sensing capabilities in atmospheric convection and turbulence. *Bull. Amer. Meteor. Soc.*, **99**, 2463–2470, <https://doi.org/10.1175/BAMS-D-17-0310.1>.
- Gottschaldt, K.-D., and Coauthors, 2017: Trace gas composition in the Asian summer monsoon anticyclone: A case study based on aircraft observations and model simulations. *Atmos. Chem. Phys.*, **17**, 6091–6111, <https://doi.org/10.5194/acp-17-6091-2017>.
- Hoell, J. M., and Coauthors, 1996: Pacific Exploratory Mission-West A (PEM-West A): September–October 1991. *J. Geophys. Res.*, **101**, 1641–1653, <https://doi.org/10.1029/95JD00622>.
- Huntrieser, H., and Coauthors, 2016: On the origin of pronounced O₃ gradients in the thunderstorm outflow region during DC3. *J. Geophys. Res.*, **121**, 6600–6637, <https://doi.org/10.1002/2015JD024279>.
- Klein, P. M., X.-M. Hu, and M. Xue, 2014: Impacts of mixing processes in nocturnal atmospheric boundary layer on urban ozone concentrations. *Bound.-Layer Meteor.*, **150**, 107–130, <https://doi.org/10.1007/s10546-013-9864-4>.
- Konopka, P., and Coauthors, 2007: Contribution of mixing to upward transport across the tropical tropopause layer (TTL). *Atmos. Chem. Phys.*, **7**, 3285–3308, <https://doi.org/10.5194/acp-7-3285-2007>.
- Lenschow, D. H., and Coauthors, 1988: Dynamics and Chemistry of Marine Stratocumulus (DYCOMS) experiment. *Bull. Amer. Meteor. Soc.*, **69**, 1058–1067, [https://doi.org/10.1175/1520-0477\(1988\)069<1058:DACOMS>2.0.CO;2](https://doi.org/10.1175/1520-0477(1988)069<1058:DACOMS>2.0.CO;2).
- Luo, Z. J., L. L. Pan, E. L. Atlas, S. M. Chelpon, S. B. Honomichl, E. C. Apel, R. S. Hornbrook, and S. R. Hall, 2018: Use of airborne in situ VOC measurements to estimate transit time spectrum: An observation-based diagnostic of convective transport. *Geophys. Res. Lett.*, **45**, 13 150–13 157, <https://doi.org/10.1029/2018GL080424>.
- McKeen, S. A., S. C. Liu, E.-Y. Hsie, X. Lin, J. D. Bradshaw, S. Smyth, G. L. Gregory, and D. R. Blake, 1996: Hydrocarbon ratios during PEM-WEST A: A model perspective. *J. Geophys. Res.*, **101**, 2087–2109, <https://doi.org/10.1029/95JD02733>.
- Müller, S., and Coauthors, 2016: Impact of the Asian monsoon on the extratropical lower stratosphere: Trace gas observations during TACTS over Europe 2012. *Atmos. Chem. Phys.*, **16**, 10573–10589, <https://doi.org/10.5194/acp-16-10573-2016>.
- National Academies of Sciences, 2016: *The Future of Atmospheric Chemistry Research: Remembering Yesterday, Understanding Today, Anticipating Tomorrow*. National Academies Press, 226 pp., <https://doi.org/10.17226/23573>.
- , 2017: The future of boundary layer observing: A workshop. National Academies of Sciences, www.nationalacademies.org/our-work/the-future-of-boundary-layer-observing-a-workshop.
- Newell, R. E., and Coauthors, 1996: Atmospheric sampling of Supertyphoon Mireille with NASA DC-8 aircraft on September 27, 1991, during PEM-West A. *J. Geophys. Res.*, **101**, 1853–1871, <https://doi.org/10.1029/95JD01374>.
- Paluch, I. R., and D. H. Lenschow, 1991: Stratiform cloud formation in the marine boundary layer. *J. Atmos. Sci.*, **48**, 2141–2158, [https://doi.org/10.1175/1520-0469\(1991\)048<2141:SCFITM>2.0.CO;2](https://doi.org/10.1175/1520-0469(1991)048<2141:SCFITM>2.0.CO;2).
- Pan, L. L., and Coauthors, 2014: Thunderstorms enhance tropospheric ozone by wrapping and shedding stratospheric air. *Geophys. Res. Lett.*, **41**, 7785–7790, <https://doi.org/10.1002/2014GL061921>.
- Patton, E. G., and Coauthors, 2011: The canopy horizontal array turbulence study. *Bull. Amer. Meteor. Soc.*, **92**, 593–611, <https://doi.org/10.1175/2010BAMS2614.1>.
- Phoenix, D. B., C. R. Homeyer, M. C. Barth, and S. B. Trier, 2020: Mechanisms responsible for stratosphere-to-troposphere transport around a mesoscale convective system anvil. *J. Geophys. Res. Atmos.*, **125**, e2019JD032016, <https://doi.org/10.1029/2019JD032016>.
- Pommrich, R., and Coauthors, 2014: Tropical troposphere to stratosphere transport of carbon monoxide and long-lived trace species in the chemical Lagrangian model of the stratosphere (CLaMS). *Geosci. Model Dev.*, **7**, 2895–2916, <https://doi.org/10.5194/gmd-7-2895-2014>.
- Sargent, M. R., J. B. Smith, D. S. Sayres, and J. G. Anderson, 2014: The roles of deep convection and extratropical mixing in the tropical tropopause layer: An in situ measurement perspective. *J. Geophys. Res. Atmos.*, **119**, 12 355–12 371, <https://doi.org/10.1002/2014JD022157>.
- Waugh, D., and T. Hall, 2002: Age of stratospheric air: Theory, observations, and models. *Rev. Geophys.*, **40**, 1010, <https://doi.org/10.1029/2000RG000101>.
- Zhang, J., and S. T. Rao, 1999: The role of vertical mixing in the temporal evolution of ground-level ozone concentrations. *J. Appl. Meteor.*, **38**, 1674–1691, [https://doi.org/10.1175/1520-0450\(1999\)038<1674:TROVMI>2.0.CO;2](https://doi.org/10.1175/1520-0450(1999)038<1674:TROVMI>2.0.CO;2).
- Zhang, Y., and Coauthors, 2016: Large vertical gradient of reactive nitrogen oxides in the boundary layer: Modeling analysis of DISCOVER-AQ 2011 observations. *J. Geophys. Res. Atmos.*, **121**, 1922–1934, <https://doi.org/10.1002/2015JD024203>.