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New Directions: Chemical climatology and assessment of atmospheric composition impacts

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Many atmospheric composition studies measure or model the concentration of X at place Y at time t , but fewer studies synthesise these measurements in the context of the full chemical environment and specific impacts. In contrast, the first systematic study of air pollution, by Victorian chemist Robert Angus Smith (1817-1884), had this explicit aim. From his

experiences with the Health of Towns Commission and as Chief Inspector of the Alkali Act (1863), Angus Smith investigated the link between atmospheric composition and human health impacts in urban areas. In his 1872 book *'Air and Rain: The beginnings of a chemical climatology'*, not only did Angus Smith coin the phrase 'chemical climatology', but he utilised methodologies recognisable today including monitoring networks with site classification, the analysis of temporal trends, and basic source apportionment (Angus Smith, 1872). Perhaps the most important legacy was his philosophy of seeking to link the atmospheric state to both causal factors and to pollution impacts. Subsequently, the term chemical climatology was used only sporadically. Recently, however, published literature containing phrases such as 'chemical climatology', 'aerosol climatology' and 'ozone climatology' have increased, but with widely varying context.

We propose that an impact-centred approach to defining chemical climatology, based on the legacy of Angus Smith, would be beneficial to establishing both relevant linkages between impacts and their drivers, and consistent syntheses of atmospheric composition studies for the research community and policy makers. To achieve this, we propose a framework that defines any climate (chemical, or otherwise, for example meteorological or political) as consisting of three elements –the **'impact'**, the **'state'** and the **'drivers'**, contained within specified spatial and temporal boundaries (Figure 1, Table 1). It is noted that some studies do fulfil the chemical climatology framework laid out here (e.g. Derwent et al., 2013). This framework is consistent with modern interpretations of a meteorological climate. For example Bryson (1997) defined meteorological climate as *'the thermodynamic/hydrodynamic status of the global boundary conditions that determine the current array of weather patterns'*. In this definition a climate 'state' determines the possible weather patterns (impacts) and is itself produced by drivers e.g. solar variability.

In the atmospheric chemical climatology context:

- **Impact** is an identified effect or metric of atmospheric composition, for which it is sought to determine the underlying contributing sources and processes. Different impacts (e.g. different metrics of the same component or of different components) are associated with different chemical climates.
- **State** is the description of the ‘what’, ‘when’ and ‘where’ of atmospheric composition producing the identified impact. This includes consideration of atmospheric constituents and their temporal and spatial variations relevant to the impact (metric), for example diurnal, annual, peak over threshold, etc. An individual chemical climate contains one state, incorporating all relevant variation.
- **Drivers** are the sources and influences on the atmospheric composition that determine the state, and hence the impact (metric). Assessment of the relative importance of each driver should explain ‘why’ and ‘how’ the composition variation detailed in the state occurs, and hence identify the dominant processes in producing instances of the impact.

The chemical climatology framework can be applied to measured or modelled data. The chemical climate is the holistic characterisation within clearly demarcated boundaries in space and time. Further, the concept of a ‘phase’ of a chemical climate (Figure 1) demarcates significant change in the drivers and state leading to significant change in the impact (metric). Phases may be identified through the segmentation of the temporal or spatial domain of a chemical climate derived using all available data, or by merging climates derived separately for a given impact over smaller temporal or spatial domains into a single climate of separate phases.

Six practical steps to define a chemical climate are summarised in Table 1, and an example template for its presentation is shown in Table 2. Step 1 identifies the impact; for example, studies link acute exposure to elevated ozone concentrations and respiratory conditions (WHO, 2006). Step 2 defines the relevant metric; e.g. maximum daily 8-h average concentration above $70 \mu\text{g m}^{-3}$, which is associated with a statistically significant increase in mortality (Amann et al., 2008). Step 3 defines the temporal and spatial boundaries to the dataset. Step 4 is the description of the state. This involves relevant temporal and spatial patterns of ozone variation above $70 \mu\text{g m}^{-3}$, e.g. diurnal and seasonal variation, and covariance with precursor molecules. Step 5 identifies drivers, for example the relative importance of local, regional and hemispheric transport, and source activities emitting ozone precursors. Step 6 assesses the presence of different phases within the chemical climate e.g. significantly different patterns of ozone metric exceedance in different regions, or significant changes to ozone precursor emissions over time. Different phases may be identified during steps 2-5 or through independent application of steps 2-5 for different spatial/temporal domains, followed by collation into a single chemical climate. Were a different impact being investigated, for example the ozone impact on vegetation (assessed by a cumulative deposition flux over a season), the state and drivers would be different, and a separate chemical climate would be derived.

Table 1 highlights the chemical climatology steps covered by four illustrative studies concerning ground-level ozone. Derwent et al. (2013) is a good recent example of a study featuring full chemical climates assessing the contribution of a driver (hemispheric baseline ozone concentrations) to different ozone impacts (vegetation and human health). Three examples of the majority of studies which assess a subset of the steps are also included in

Table 1. WHO (2006) assess the health impact of ozone and define a relevant metric (steps 1 and 2), but do not evaluate the state and drivers of ozone variation in particular locations; Malley et al. (2014) describe changes in ozone variation at rural sites across Europe (steps 3 and 4), but do not link to ozone impacts or causal drivers; Gerasopoulos et al. (2006) assess the state and drivers of ozone variation at Finokalia, Crete (steps 4 and 5), but do not link this variation to ozone impacts, nor evaluate the temporal and spatial representativeness of ozone variation at the location. Covering a subset of the chemical climatology steps is not a shortcoming of studies, and neither should every investigation aim to cover every step in the chemical climatology framework. However, increased awareness of the steps within the framework covered by isolated studies means that they can be combined to produce full impact-led chemical climate assessments focussing on relevant local, regional and global scale issues. This would better facilitate consideration of impact mitigation strategy development where needed. A standard output from chemical climate studies (Table 2) summarises the statistical features of the chemical climate, as well as the temporal and spatial boundaries and scientific uncertainties. This could allow collation and linkage between chemical climates.

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Table 1: Chemical climatology framework: Component Steps and a few example studies identifying which component steps were described

Step	Description	Example chemical climatology	Example studies			
		Ozone	<i>Gerasopoulos et al. (2006)</i>	<i>WHO (2006)</i>	<i>Derwent et al. (2013)</i>	<i>Malley et al. (2014)</i>
1	Identify impact	Human health; Vegetation damage		✓	✓	
2	Define relevant chemical climate metric(s) for the impact	Sum of means over 35 ppb (SOMO35); Accumulated ozone over 40 ppb (AOT40)		✓	✓	
3	Define the chemical climate's temporal and spatial boundaries	Representivity of time period and location			✓	✓
4	Describe the chemical climate state	Statistical analysis of measured/modelled dataset	✓		✓	✓
5	Identify the chemical climate driver(s)	Relative contribution of meteorology, source apportionment, atmospheric chemistry	✓		✓	
6	Assess for phases within the chemical climate	Significant temporal/spatial changes in impact severity			✓	

Figure 1: An illustration of the chemical climatology framework. For a particular chemical climate description, only a single phase might be identified.

