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NOTES AND CORRESPONDENCE

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Low number concentration of ice nucleating particles in an aged smoke plume

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Smoke from forest fires in the western part of North America reached the High Altitude Research Station Jungfraujoch, Switzerland, at the beginning of September 2017. Number concentration of ice nucleating particles (INPs) active at −15 °C or warmer decreased by about half during its passage. This is different to observations of enhanced INP concentrations in fresh plumes from forest fires. We hypothesise that INPs initially present in a smoke plume are lost or deactivated during long-range transport, while components of smoke capable of deactivating INPs originally present or mixed later into the plume continue to remain active across a longer distance.

KEYWORDS
coating, deactivation, forest fire, ice nucleating particles, smoke plume

1 | INTRODUCTION

Dozens of forest fires had shrouded large parts of the Pacific Northwest, USA, and western Canada at the beginning of September 2017 in a cloud of smoke and ash (Stevens, 2007). A large fraction of the smoke cloud detached, crossed North America and the Atlantic, and arrived after about 4 days in western Europe, as satellite data have shown (NASA Earth Observatory, 2017). One of the most comprehensive measurement programmes of *in situ* trace gas and aerosol properties at a mountain site worldwide is run at the High Altitude Research Station Jungfraujoch (3,580 m above sea level (a.s.l.)) in Switzerland. While the station is predominantly exposed to pristine lower free troposphere air masses, several parameters clearly indicated the presence of smoke for a period of approximately 2 days, from 7 September in the evening until 9 September around noon (Figure 1). We took this rare chance to investigate the long-distance impact of forest fires on ice nucleating particles (INPs).

Previous observations in the USA have shown that forest fires can regionally enhance INP number concentrations above background (Prenni *et al.*, 2012; McCluskey *et al.*, 2014), though not all fuels produce INPs (Petters *et al.*, 2009) and smouldering fires are a negligible source of INPs (Prenni *et al.*, 2012). However, flaming fires, such as the ones having caused the plume we sampled, can double or triple INP number concentrations regionally and measurably enhance them even 1,600 km downwind (Prenni *et al.*, 2012). Upon further transport, the sign of the impact may change. For example in October 2004, numerous fires in Siberia could have led to the substantial decrease in INP number concentration registered 3–4 days later above Alaska, where an increase in the fraction of carbonaceous particles was observed simultaneously (Prenni *et al.*, 2009).

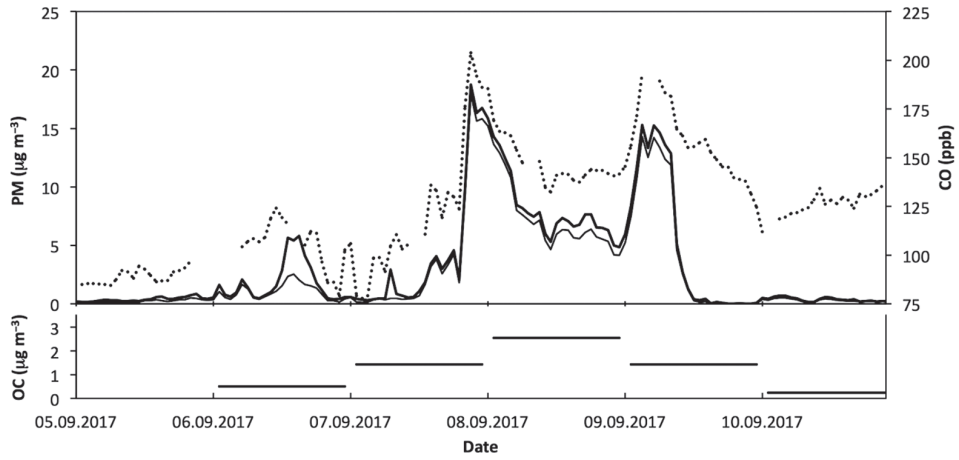

FIGURE 1 Upper panel: Six-day time series of PM_{10} (thick line), PM_1 (thin line), and CO (dotted line) at Jungfraujoch. Enhanced concentrations of PM from 7 September 2017 in the evening until 9 September around noon were associated with an average Scattering Ångström Exponent (SAE) of 2.0 and an Absorption Ångström Exponent (AAE) of 1.3, indicating an elemental/organic carbon (EC/OC) mixture (Cazorla *et al.*, 2013). The SAE value clearly distinguishes the event from a Saharan dust episode, when SAE is around 1.0. The ratio of PM_{10} to PM_1 would also be much larger for dust episodes. Lower panel: OC concentrations derived from PM_{10} filters sampled over 24 h

2 | METHODOLOGY

A comprehensive set of aerosol physical properties and trace gases are continuously measured at Jungfraujoch (7°59′02″E, 46°32′53″N, 3580 m a.s.l.) as part of the Swiss contribution to the World Meteorological Organization (WMO)'s Global Atmosphere Watch (GAW) programme and the operation of the Swiss National Air Pollution Monitoring Network NABEL (Bukowiecki *et al.*, 2016; Steinbacher *et al.*, 2017). Next to the continuous analysis, integrated samples of particulate matter <10 μm (PM_{10}) are collected at a rate of 30 m^3/h on quartz-fibre filters (140 mm effective diameter) for 24 hours starting midnight. We analysed sections of these filters for their content of INPs active at −15 °C or warmer (INP_{-15}). From each filter, 54 punches (1 mm diameter) were immersed in the same number of 0.5 mL Eppendorf tubes with 0.1 mL ultrapure water and exposed in a cold bath to a temperature of −7 °C, declining over the course of 25 min to −15 °C. The number of frozen tubes was observed visually at −15 °C. The method is described in more detail in Conen *et al.* (2012; 2015). Apart from the 3 days with enhanced markers of smoke (7–9 September 2017) we also analysed filters of the 2 preceding days, and of the day following the event. For comparison, we include data from background conditions during summer ($PM_{10} < 2$ $\mu g/m^3$) and two Saharan dust events observed during the summers of 2016 and 2017. Saharan dust events are clearly recognisable at Jungfraujoch in the ochre colouring of PM_{10} filters, in back-trajectories and in aerosol parameters (Collaud Coen *et al.*, 2004; Conen *et al.*, 2015). Number concentrations of particles were determined with an optical particle counter (TSI 3330, TSI Incorporated), which measures the number size distribution in 16 size channels between 0.3 and 10 μm (optical diameter), as well as with a scanning mobility particle sizer (SMPS: Wiedensohler *et al.*, 2012) in the size range 16–591 nm (mobility diameter).

3 | RESULTS AND DISCUSSION

The average number concentration of particles larger than 0.5 μm in diameter ($n_{0.5}$) increased during the smoke event by about a factor of 20. This increase in $n_{0.5}$ was dominated by an increased particle number between 0.5 and 0.6 μm optical diameter as a result of strongly increased upper tail of the accumulation mode. The modal diameter increased from 90 to 200 nm (mobility diameter) and the particle number between 100 and 500 nm increased from <100 to 300–600 cm^{-3}. In contrast, number concentrations of INP_{-15} were smaller, only about half as large, during the smoke episode compared to the 3 days bracketing it. Compared to other background values in summer, INP_{-15} concentration was not particularly high before and after the event (Figure 2). Observed INP_{-15} concentrations during background conditions were between the two empirical parametrizations proposed by DeMott *et al.* (2010; 2015) for INPs as a function of $n_{0.5}$ and temperature. The earlier parametrization is based on a large number of field observations from a variety of locations on Earth; the later parametrization additionally includes laboratory data. The later parametrization is explicitly for the immersion freezing ability of mineral dust particles. It is in good agreement with our observations of Saharan dust events. However, the smoke event was clearly below the lower prediction (Figure 2). This implies that the upper tail of the accumulation mode size distribution, which dominated $n_{0.5}$ during the smoke event, gave an under-proportional contribution to INP_{-15} compared to INP_{-15} number fraction relative to $n_{0.5}$ typically observed during background and Saharan dust conditions at Jungfraujoch.

Background aerosol particles mixed into the smoke plume probably constituted the majority of INP_{-15} at Jungfraujoch during the smoke event, because the number concentration of INP_{-15} during the event was only half the background value before and after the event. Hence, it is unlikely that the smoke itself contained relevant numbers of INP_{-15} when it reached

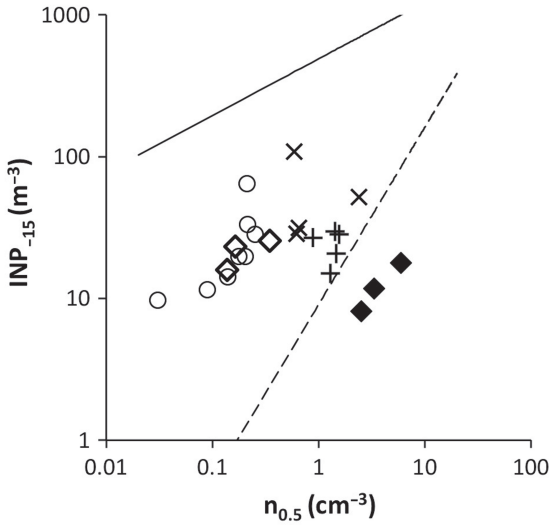

FIGURE 2 Number concentration of INP_{-15} in relation to particles larger than 0.5 μm ($n_{0.5}$) at Jungfraujoch during the passage of smoke from distant forest fires (filled diamonds). The values 2 days before, and the day after the smoke event, (open diamonds) fall into the cluster of other background values (circles; $PM_{10} < 2\ \mu g/m^3$). Data of two Saharan dust events during summer are added for comparison (×: 2016; +: 2017). The two lines indicate parametrizations of INP_{-15} as a function of $n_{0.5}$ (continuous line: DeMott *et al.*, 2010; dashed line: DeMott *et al.*, 2015). For control, we analysed from two filters in each cluster (background, Saharan dust events, smoke event) the 5 mm wide fringe, where no air has passed through. These upper estimates of blank values (Conen *et al.*, 2012) were on average 8% of the sample values shown in the plot

Jungfraujoch, unless it was diluted less than 1:1 with background air, which is unlikely after the travelled distance. On the contrary, some INP_{-15} in the background aerosol seem to have been deactivated by mixing with smoke. Organic aerosol constitutes the largest mass fraction of aerosol (PM_1) in forest fire plumes (Kondo *et al.*, 2011) and gas phase diffusion efficiently redistributes its semi-volatile components between aerosol particles in the lower troposphere (Marcolli *et al.*, 2004). Condensation of such compounds onto background particles could have deactivated some of their ice nucleation-active sites (Möhler *et al.*, 2008; Primm *et al.*, 2017). Similarly, secondary organic aerosol particles generated in a smoke plume can coagulate with background particles and cover large parts of their surface (Deboudt *et al.*, 2010). In our observation, the deactivating compounds must have been water-insoluble, otherwise they would have been removed from the ice nucleation active sites during the immersion freezing assay. Alternative explanations for the observed decrease in INP concentrations during the smoke event include their deactivation by compounds that were not emitted by the forest fires, sedimentation of activated INP with ice crystals prior to the arrival of the smoke-affected air mass at Jungfraujoch, and initially reduced number concentrations of background INP_{-15} in the smoke-dominated air masses.

Previous observations revealed the potential of forest fires to regionally increase the number concentration of INPs active at temperatures colder than −15 °C (Petters *et al.*, 2009; Prenni *et al.*, 2012; McCluskey *et al.*, 2014), although the ratio of INP to $n_{0.5}$ is smaller in fresh smoke than in background air (McCluskey *et al.*, 2014). Our opportunistic study of INP_{-15} during the passage of aged smoke also shows a small ratio of INP to $n_{0.5}$, but no increase in INP concentrations. Absolute concentration of INP_{-15} was smaller in the smoke-affected air mass than in background air, similar to the observation made by Prenni *et al.* (2009) in Alaska. Based on these initial pieces of evidence we propose the following working hypothesis: INPs initially present in smoke plumes are lost or deactivated during long-range transport, while components of smoke that deactivate INPs, also in background air masses, are transported further.

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