# Formation of Environmentally Persistent Free Radicals During the Atmospheric Ageing of Pollen

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**Abstract**

Environmentally persistent free radicals (EPFRs) can generate reactive oxygen species (ROS) and are thus considered toxic. The primary and secondary sources of EPFRs, especially in the context of atmospheric particulate matter, remain uncertain. Here, we present the first measurements of EPFRs in tree pollen and explore their formation and destruction in the atmosphere using a combination of environmental sampling and laboratory experiments. First, we present results from a Spring 2023 sampling campaign which collected deposited particulate matter and partitioned organics, referred to as grime. Pollen comprised most of the grime mass and total EPFR signal when isolated from the smaller (< 10 μm) grime components. Over the duration of the sampling, the number of EPFRs normalized to the mass of pollen increased, suggesting that the EPFRs from the pollen were being formed via dark secondary processes post-deposition. In the lab, irradiation of the pollen-covered surface increased the total number of EPFRs, showing that photoaged pollen could generate more ROS. Second, we show that tree pollen (*Quercus velutina*, *Quercus rubra* and *Betula Pendula*) shows an endogenous EPFR signal, located on the outside of the pollen, as opposed to the inside which is inaccessible to the atmosphere. We use this suite of pollen to explore the influence of irradiation, atmospheric oxidation (ozone exposure), and pollen rupture to discuss the role of atmospheric processing on total EPFRs in pollen. This study highlights the contribution of pollen to atmospheric EPFRs, and the need to consider the influence of atmospheric processing on the total number of EPFRs on pollen. As the average temperature around the globe increases, pollen seasons are starting earlier and lasting longer, which increases the EPFR burden from a previously unknown source.

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