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Invited review article

A review of observations of organic matter in fogs and clouds: Origin, processing and fate

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ABSTRACT

While fog and cloud composition has been studied for decades, most of the research was limited to inorganic species and fog acidity. Recently the focus has shifted towards organic matter in the atmospheric aqueous phase of fogs and clouds: its origin, reactivity and fate. An impressive number of fog and cloud chemistry observational studies have been performed over the last decade throughout the world. In the present work we will review the state of knowledge of atmospheric organic matter processing by fogs, with a focus on field observations. We start by reviewing observational studies in general and then discuss our knowledge on the occurrence of organic matter in fogs, its solubility, characterization and molecular speciation. Organic carbon concentrations can vary widely from approximately 1 mg C/L in remote marine environments to more than 100 mg C/L in polluted radiation fogs, accounting for a substantial part of fogwater solutes. The carbonaceous material can enter the droplets from the gas and particle phase and the scavenging behavior of fogs will be detailed. Observational studies showed evidence of aqueous phase transformation of organic material, in particular secondary organic aerosol (SOA) generation, in fog. Recent observations of biological material in fog suggest also an impact of biological processing within the droplets on fog organic matter. The review will end with a discussion of the impact of fog on the deposition fluxes of organic material and hence its atmospheric lifetime.

Contents

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1. Introduction

While the earliest measurements of fog and cloud composition were reported in the middle of the 20th century (Houghton, 1955), observational research on this topic accelerated beginning in the 1980s with the discovery of acid fog (e.g., Waldman et al., 1982). Much of the research in the 1980s focused on fog and cloud acidification, including sulfur oxidation processes (e.g., Munger et al., 1983; Barrie, 1985; Hoffmann, 1986), with additional work examining deposition of water and cloud-borne solutes from cloud interception on forested hillsides (Lovett, 1984; Fuzzi et al., 1985; Waldman et al., 1985; Schemenauer, 1986). This was also a period when research into acid precipitation received considerable attention in the U.S. and in Europe. In the 1990s research shifted more toward understanding processing of air pollution by clouds and fogs (Facchini et al., 1999). This included increased attention to finer details of cloud/fog composition (e.g., examining differences in composition with droplet size) (Collett et al., 1993a, 1994; Bator and Collett, 1997) and studies of the net effect of fogs and clouds on airborne fine particle concentrations, through aqueous phase chemical production that increased fine particle mass released by evaporating fogs/clouds and fine particle scavenging and deposition via fog drop sedimentation or incorporating of cloud drops into ice crystals via riming in mixed phase clouds (e.g., Fuzzi et al., 1992; Collett et al., 1991, 1993b; Wobrock et al., 1994). A number of researchers also grew increasingly interested in aqueous phase photochemistry in cloud and fog drops (Erel et al., 1993; Anastasio et al., 1994; Zuo and Hoigne, 1994). Starting in the 1990s, but with increased emphasis in the 2000s, fog and cloud studies addressed organic matter, its characterization, sources and processing. Since that time, interest has increased, in particular, in the formation of secondary organic aerosol through fog and cloud processing of volatile organic precursors. While much of the early work on fog and cloud chemistry was conducted in the U.S. and Europe, in the last 15 years fog and cloud chemistry observational studies became more global and a small, but active international research community emerged. Since 1998 a series of international conferences on fog, fog collection, and dew has been held at locations in N. America, Africa, S. America, Europe, and Asia. Special journal and book issues published in conjunction with some of these meetings (e.g., Collett et al., 2002; Gultepe et al., 2007; Berkowicz et al., 2008; Eugster, 2008; Gultepe, 2012; Bendix et al., 2012) contain many papers that provide insight into some of the topics of investigation in recent decades.

In the present manuscript we will review the state of knowledge concerning atmospheric organic matter processing by fogs, with a focus on field observations. The term fog will be used from here forward to refer to radiation fogs, but also other types of fogs as well as clouds that touch the ground, assuming that any cloud that touches the ground is considered a fog. In terms of chemistry and chemical processing there are many similarities between fogs and clouds, although they can be substantially different in terms of atmospheric conditions in which they form or in terms of microphysical processes. Finally, we will focus mainly on scientific results published over the last decade.

Fig. 1 illustrates the impacts of fog processing on atmospheric chemistry and, in particular, on atmospheric organic matter in both the gas and particle phases. In this review we will first summarize recent fog and cloud chemistry observational studies. In a second section we discuss available observations of organic matter concentrations in fogs, its solubility, characterization and molecular speciation. In a third section the state of knowledge of the scavenging of gas and particle phase organics by fogs will be discussed. The fourth section will provide an overview of organic cloud chemistry research in recent years, while fifth section will introduce the relatively new area of biological material in fog and discuss its contribution and impact on fog organic matter. The last section of this review will discuss deposition fluxes of organic matter by fogs.

1.1. Recent developments in observational fog studies

Many recent observational studies of fog chemistry have been conducted; these are briefly summarized here. While in the 1980s and 1990s fog chemistry observations were mostly made in Europe and North America, with a few locations in Japan and elsewhere in Asia, since 2005 there has been a strong increase in observational studies in Asia, in particular in China (Gao et al., 2012; Guo et al., 2012; Li et al., 2011a; Liu et al., 2012; Liu and Wang, 2005; Shen et al., 2012; Wang et al., 2011a, 2011b; Niu et al., 2010 and references therein), but also in Japan (Aikawa et al., 2007a, 2007b; Matsunaka et al., 2005; Watanabe et al., 2006, 2010, 2011), Taiwan (Sheu and Lin, 2011), South Korea (Kim et al., 2006) and India (Kaul et al., 2011; Lakhani et al., 2007; Safai et al., 2009).

In Europe, recent fog chemistry observations were reported from Poland (Błaś et al., 2008, 2010) and the Czech Republic (Fisak et al., 2008; Zapletal et al., 2007) while several large field studies were conducted in Germany to investigate cloud and fog processing of both inorganic and organic pollutants (Herrmann et al., 2005a; van Pinxteren et al., 2005; Harris et al., 2013). Other well established measurement locations continued to report observations including the Puy de Dôme in France (e.g., Marioni et al., 2004) and the Po Valley in Italy (e.g., Decesari et al., 2000). A recent large field campaign (PARISFOG) was conducted in France and results are forthcoming (Haeffelin et al., 2010).

In North America, fog studies continued in the Central Valley of California (Herckes et al., 2007a, 2007b; Collett et al., 2008; Mazzoleni et al., 2010; Ehrenhauser et al., 2012) as well