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Presence of endocrine disruptors in freshwater in the northern Antarctic Peninsula region

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ABSTRACT

The increasing human presence in Antarctica and the waste it generates is causing an impact on the environment at local and border scale. The main sources of anthropic pollution have a mainly local effect, and include the burning of fossil fuels, waste incineration, accidental spillage and wastewater effluents, even when treated. The aim of this work is to determine the presence and origin of 30 substances of anthropogenic origin considered to be, or suspected of being, endocrine disruptors in the continental waters of the Antarctic Peninsula region. We also studied a group of toxic metals, metalloids and other elements with possible endocrine activity. Ten water samples were analyzed from a wide range of sources, including streams, ponds, glacier drain, and an urban wastewater discharge into the sea. Surprisingly, the concentrations detected are generally similar to those found in other studies on continental waters in other parts of the world. The highest concentrations of micropollutants found correspond to the group of organophosphate flame retardants (19.60–9209 ng L^{-1}) and alkylphenols (1.14–7225 ng L^{-1}); and among toxic elements the presence of aluminum (a possible hormonal modifier) (1.7–127 μg L^{-1}) is significant. The concentrations detected are very low and insufficient to cause acute or subacute toxicity in aquatic organisms. However, little is known as yet of the potential sublethal and chronic effects of this type of pollutants and their capacity for bioaccumulation. These results point to the need for an ongoing system of environmental monitoring of these substances in Antarctic continental waters, and the advisability of regulating at least the most environmentally hazardous of these in the Antarctic legislation.

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Abbreviations: BeP, Benzylparaben; BPA, Bisphenol A; BT, *1H*-Benzotriazol; CAS, Chemical Abstracts Service; DES, Diethylstilbestrol; DDT, Dichlorodiphenyltrichloroethane; E1, Estrone; E1-3s, Estrone-3-sulfate; E1-3g, Estrone-3-glucuronide; E2, 17-β-estradiol; E2-17g, Estradiol-17-glucuronide; E3, Estriol; E3-3s, Estriol-3-sulfate; E3-16g, Estriol-16-glucuronide; EDCs, Endocrine-disrupting compounds; EE2, Ethinylestradiol; EtP, Ethylparaben; HCB, Hexachlorobenzene; HCH, Hexachlorocyclohexane; MeP, Methylparaben; NP, Nonylphenol; NP_1EC, Nonylphenol monocarboxylate; NP_1EO, Nonylphenol monoethoxylate; NP_2EO, Nonylphenol diethoxylate; OP, Octylphenol; OP_1EC, Octylphenol monocarboxylate; OP_1EO, Octylphenol monoethoxylate; OP_2EO, Octylphenol diethoxylate; POPs, Persistent organic pollutants; PET, Polyethylene terephthalate; PrP, Propylparaben; PVC, Polyvinyl chloride; TBEP, Tris (butoxyethyl) phosphate; TCC, Triclocarban; TCPP, Tris (1-chloro-2-propyl) phosphate (TCPP); TCEP, Tris (2-cloroethyl) phosphate; TCS, Triclosan; TT, Tolytriazol; US EPA, United States Environmental Protection Agency; WHO, World Health Organization; WWD, Wastewater Discharge

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1. Introduction

Antarctica is the last continent to be settled by humans. Its distance from populated areas and extreme climate has maintained it relatively isolated from sources of pollution. Since the coming into effect of The Antarctic Treaty (23 June, 1961), the entire continent has been dedicated to science, with the adoption of the Protocol on Environmental Protection, also known as Madrid Protocol (signed in 1991 and entry into force in 1998), and all activities that could potentially alter its original condition have been restricted or prohibited.

The Antarctic environment is among the most extreme on Earth due to the intense cold and special light conditions, which restricts animal and plant development. In spite of this, numerous ecosystems are particularly activated during the brief austral summer. This season is also the period of maximum scientific activity and the greatest influx of tourists. There are currently over 100 Antarctic scientific bases, shelters and other human facilities on the continent, and recent years have routinely seen the arrival of as many as 30,000 tourists annually (IAATO, 2015).

The greatest numbers of facilities and visitors are clustered in the Antarctic Peninsula region, due to easier access and less extreme climate conditions. The environmental conditions in this area favor greater development of life, and it has also suffered the fastest rate of warming in the whole southern hemisphere in recent decades (Turner et al., 2005).

The Madrid Protocol (1998) has regulated human activity in Antarctica and sought to prevent the entry of extraneous elements, the dispersion of pollutants and their effects in the environment. Despite this, the presence of humans in Antarctica has caused an impact at the local level in addition to others of a global nature (Bargagli et al., 2005; Bergstrom et al., 2006; UNEP, 2002). In fact some pesticides such as dichlorodiphenyltrichloroethane (DDT) and similar, in combination with other persistent organic pollutants (POPs) such as hexachlorobenzene (HCB) and hexachlorocyclohexane (HCHs) have been detected in Antarctic marine biota and in other abiotic matrices when no applications have ever taken place on this continent (Sladen et al., 1966; UNEP, 2002).

A particular feature of the study area is that during the winter the hydrological system remains frozen (Turner et al., 2005), and the pollutants are therefore immobilised in the soil or solid water (buried ice, snow, permafrost) (Michel and Van Everdingen, 1994). The rise in temperature during the summer has the effect of activating the water cycle in certain sectors and causing the entry in circulation of large amounts of liquid water, the mobilization of nutrients, and –in the case of areas impacted by human activity– the mobilization of pollutants through water courses and aquifers.

The hydrological dynamics in this environment is linked to the thaw and to the development of the active permafrost layer, creating a connection with varying degrees of intensity between the groundwater, streams and ponds (French, 2013). During the summer, the active layer accumulates liquid water that circulates over the permafrost –which constitutes the impermeable layer– and joins the main flow (Moreno et al., 2012). Another notable feature of the water flow in the streams in this area is its daily –and even hourly– variability, conditioned by factors such as air temperature, insolation rate and the occurrence of precipitation.

The main sources of pollution from local anthropic activities in the soil, air, water, snow and biota of Antarctica include the burning of fossil fuels, waste incineration, accidental spillage and wastewater discharge (Bargagli, 2008). As of 2009, over one third of the wastewater from the permanent bases and over two thirds of the Antarctic summer stations have no wastewater treatment systems (Gröndahl et al., 2009). The existing treatment systems are often unable to cope with the increase in effluents that occurs in summer (Gröndahl et al., 2009), and the presence of biologically active substances such as personal hygiene products and steroid hormones has been reported in the coastal environment around the Antarctic stations (Emnet et al., 2015). These compounds, in conjunction with other substances that may occur naturally in the Antarctic environment (toxic elements), might have adverse effects on organisms.

For many years these micropollutants have been disregarded as an environmental risk by classical toxicology, since their environmental concentrations are below the toxic thresholds obtained from standardised dose-response curves. However, it has long been known that toxicants can exert biological activity well below their toxic threshold concentration (Evenari, 1949; Calabrese and Blain, 2009). These sublethal effects are not directly associated with adverse events, but frequently result in the disruption of cell communication or endocrine regulation systems. The subtle disruption of an organism's homeostasis at critical points of the life cycle can lead to biological failure even though the toxicant exerts no apparent toxicity (Deblonde et al., 2011).

Advances in analytical chemistry now permit the detection of a wide array of substances in the μg and even ng per litre range. Hundreds or even thousands of substances derived from human presence, activity or industry have been detected in the environment, swelling the ranks of the so-called emerging pollutants. Water treatment technologies are not designed to eliminate them, meaning that anthropogenic microcontamination of freshwaters is now a widespread reality and has been reported all over the planet (Brausch and Rand, 2011; Kasprzyk-Hordern et al., 2008; Giordano et al., 2009; Kim et al., 2007; Kuster et al., 2008; Kolpin et al., 2002; Pal et al., 2010; Wang et al., 2012).

The United Nations Environment Programme identified around 800 chemicals with known or presumed endocrine disrupting ability (Bergman et al. 2012), and the European Union has compiled a two-tier list of substances inducing a clear (Category 1, >190 substances) or suspected disruptive activity (Category 2, >120 substances) (Petersen et al., 2007) which have been associated to altered metabolism and behavior, developmental abnormalities and a decrease in fertility affecting all aquatic trophic levels (fish, algae, amphibians, invertebrates, and so on) (Pal et al., 2010). Their link with human metabolic, immune, neurological and behavioral disorders (Bergman et al. 2012; Lyche et al., 2011) is of particular concern due to the high exposure through food, drinking water and personal hygiene products, especially during pregnancy, infancy and old age (Bergman et al. 2012; Llorca et al., 2010; Mantovani, 2015).

The US Environmental Protection Agency (US EPA) defines an endocrine disrupter as "an exogenous agent that interferes with the production, release, transport, metabolism, binding, action or elimination of natural hormones in the body responsible for the maintenance of homeostasis, reproduction, development and/or behavior" (Kavlock et al., 1996). This is a highly heterogeneous group of substances (natural and synthetic, organic and inorganic) with different chemical structures and used in almost every human activity, from industry to personal hygiene products, and including pharmaceuticals and food preservatives. Alkylphenols, bisphenol A, chlorinated aromatic phenolics (i.e. disinfectants), parabens (i.e. preservatives) and organophosphorous compounds (i.e. flame retardants) are among the most suspicious substances. Natural human and synthetic estrogens are also included within this type of micropollutants. Other inorganic substances such as certain heavy metals (Hg, Al, Pb, Cd) and metalloids (As) may also be able to alter metabolism and hormonal activity (Adams et al., 2014; Ciarrocca et al., 2012; Correia et al., 2010; Ettinger et al., 2014; Liu et al., 2014; Padilla et al., 2010; Xu et al., 2012). Concretely, the work of Dyer (2007) reported that some heavy metals mimic the biologic activity of steroid hormones, including androgens, estrogens, and glucocorticoids, and could have an impact on mammalian reproductive systems.

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