TRANSBOUNDARY AIR POLLUTION

Franco DiGiovanni and **Philip Fellin**
Airzone One Ltd., 222 Matheson Boulevard East, Mississauga Ontario, Canada

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Summary

A broad range of pollutants have been described that share the common phenomena of transboundary transport. It has been seen that transboundary transport occurs either because the pollutants have very low deposition velocities (as in the case of Arctic haze constituents or fine particulate matter (PM)), or an extended period of time is required for the pollutant to develop from the precursor compounds (smog, acid rain) or are chemically inert (mercury) or go through a multi-hop pathway as in the case of persistent organic pollutants (POPs). In general, the smog and acid rain issues have had the longest history of study and are generally better understood. More recently issues of POPs and mercury have come to the fore, and research is in early development, possibly because their effects are more insidious.

There are two major challenges with transboundary pollutants. The first is the international co-operation required to deal with them. Fortunately, in recent years, the political climate has become more attune to international co-operation to alleviate transboundary air pollution (e.g., North American Free Trade Association – Commission of Environmental Co-operation, rationalization of EU environmental regulations, UN CLRTAP). Other factors aide these political decisions, which must balance social and economic considerations against environmental, such as the recognition of co-benefits of emissions reductions where certain pollutants have multiple effects (e.g. NO_x in acid rain and smog issues).

The second challenge involves providing the appropriate data upon which reduction decisions can be made. These data are generated by the scientific monitoring and modeling investigations described in this paper. For transboundary pollutant problems, the need to co-ordinate international networks of monitoring stations has been successfully met in Europe (EMEP) and in North America (Environment Canada 1998) and modeling efforts have often been coordinated, although to a greater extent in Europe under the EMEP Program than in North America. Given the complexity of the vertical and horizontal structure of the atmosphere, and the large range of scales atmospheric motion affecting transboundary air pollution, the types of complex modeling efforts require are perhaps best accomplished by international co-operation.

1. Introduction

Pollutants emitted from natural or anthropogenic sources to the atmosphere may be advected over distances of several to hundreds of meters (micro-scale), 1000's of meters to hundreds of kilometers (meso-scale) or hundreds to 1000's of kilometers (macro-scale) (Oke 1978). The upper end of macro-scale dispersal describes global dispersal patterns and is also referred to as ultra-long range transport. When airborne contaminants cross geopolitical boundaries or migrate across several geographic zones, the pollution is designated as transboundary even if the physical distance of the boundary from the emitting pollutant source is quite short.

Many transboundary issues trace their origin to releases from specific sources impinging directly upon receptors in other jurisdictions. For example, a smelter in Trail, British Columbia, released SO_2 from a tall stack that impinged directly on sites in Washington State, USA, under specific atmospheric conditions. This issue was the subject of protracted negotiations from the 1930s to the 1950s when it was resolved by treaty negotiated through the International Joint Commission (IJC). The "Superstack" in Sudbury, Ontario, a 335 m chimney erected to disperse SO_2 emissions from a nickel smelter in the late 1960s, became the single largest source of SO_2 emissions in North America, initially at about 5 000 Tonnes/annum (currently reduced to less than 500 Tonnes/annum). Emissions were tracked as far as lower New York State in the USA. Many more examples are available from all over the world of transboundary pollution problems of this type.

Over the last thirty years or so, however, transboundary air pollution has become synonymous with broader, more complex issues incorporating contributions from many sources and vast regions, complex atmospheric processes and multiple chronic effects on receptors that are more difficult to detect and define unambiguously. The latter view of transboundary air pollution is discussed in this article drawing upon research experiences and regulatory and environmental issues in North America, similar work in Europe and broader international efforts including the United Nations Environment Program.

In this paper we begin with a brief overview of atmospheric processes that affect air pollution transport. The rest of the paper will describe the major transboundary pollutants. Specifically, we will describe their sources, aspects of their long-range transportation, and their deposition or incorporation into the ecosystem.

Appropriate reduction strategies require knowledge of the emissions, dispersal and deposition processes, as well as an understanding of atmospheric processes and use of that knowledge to identify significant sources. This knowledge and data is gained by monitoring of atmospheric pollutants, often utilizing networks of monitoring stations, and computer-based models of atmospheric dispersal and chemical processes that allow prediction of the affects of suggested reduction methods. Such models require monitoring data for validation, and thus monitoring and modeling studies are often conducted in parallel.

To effectively manage a transboundary pollution issue, the source-receptor relationship is an important one to elucidate. In other words, it is important to know how much deposition at location "y" occurs as a result of emissions from location "x" and, moreover, to understand if a linear or non-linear relationship exists between source emissions and receptor deposition. Both these factors are crucial to applying appropriate controls, and are especially important considerations in transboundary transport where an appreciable disjoint may develop between variations in the pollution emission and variations in atmospheric concentrations of the pollutants at the receptor. It is the relationship between emissions and final deposition of the pollutant that computer models attempt to simulate. Successful simulations allow the models to be used in a predictive mode to assess the success of various proposed reduction strategies. Accordingly, the last two sections of each pollutant description will

describe examples of investigative techniques and reduction strategies that may have been implemented or are planned.

2. Background on the Atmosphere and Dynamics

When natural or anthropogenic pollutants are emitted from their source, air currents carry the pollutants, dilute them, and expose them to varying environmental conditions or other chemicals in the atmosphere, before the pollutants are either chemically transformed or deposited to the earth's surface (land or water) through dry or wet deposition processes. Therefore, it is essential to understand the atmospheric structure and dynamics that control these air movements. A description is given based upon issues relevant to long-range transport.

2.1. Vertical Structure of the Earth's Atmosphere

The vertical structure of the earth's atmosphere is classified by the thermal gradient. The bottom layer, where the air temperature generally decreases with increasing altitude, is known as the troposphere. The height of the troposphere varies from approximately 16 km over the Tropics to approximately 9 km over the Polar region. Above the troposphere, between approximately 12 and 50 km, the stratospheric layer contains much of the ozone that protects the earth from sun's UV radiation.

Within the troposphere, the vertical structure is further sub-divided into the surface layer, the planetary boundary layer (PBL) and the free atmosphere. It is within the PBL that most of the "weather," that affects pollutant transformation and dispersal, occurs. The surface layer is defined as the layer with approximately constant shear stress (a measure of the drag of the earth's surface on the atmosphere) with increasing height where the winds are determined by the nature of the surface and vertical gradients of temperature. This layer may extend from 50 - 100 m above the earth's surface. The rest of the planetary boundary layer extends up to approximately 500 – 3000 m and is a region of transition wherein shear stresses are variable and winds are determined by horizontal pressure gradients, Coriolis and surface friction forces and also vertical temperature gradients. The variation of forces with increasing height causes wind directions to vary as height increases. Above the planetary boundary layer, the balance of the troposphere is made up by a region called the free atmosphere where air motions are governed by the horizontal (synoptic) pressure gradients and the Coriolis force and flow is quite laminar with winds of a high velocity.

The top of the boundary layer is marked by a temperature inversion, that is, a layer above which environmental temperatures increase. Such a "capping" inversion usually traps pollutants released at the earth's surface within the boundary layer since vertical movements are subdued. Turbulence and convective activity within the PBL causes pollutants to be mixed within this layer. Because the height of the PBL varies, and especially is lower at night, highly buoyant pollutants, released from an industrial smoke stack, for example, or from other elevated point sources may penetrate the inversion and invade the free atmosphere where the potential for long-distance dispersal is greatly enhanced. Also, pollutants released into the nocturnal boundary layer (NBL) can result in elevated atmospheric concentrations because there is less

depth available for dilution of the pollutant. The interested reader is directed to Stull (1997) and Lutgens and Tarbuck (1986) for further details.

2.2. Horizontal Structure of the Earth's Atmosphere

Wind flow over the earth's surface is affected by phenomena of varying length and time scales. Pollutants released into the atmosphere, and which take part in transboundary pollution, are influenced by motions covering virtually the entire spectrum of atmospheric motions. At the larger scales, the influences are controlled by large scale global circulation patterns, and at the smaller scales by small scale turbulence and air viscosity effects.

Air masses on the earth's surface have distinct characteristics of temperature and humidity derived from the characteristics of the surface over which it resides. As a result, meteorologists have classified air masses according to their sources region. Thus the Arctic air mass has characteristics derived from the Arctic region, the Polar air mass is characteristic of temperate regions and the Tropical air mass has climatic features typical of the Tropics. The general circulation over the earth is such that warmer air from the topics moves towards the poles and cold air from the poles moves towards the equator. The flow patterns are modified by the rotation of the earth, and the three-cell meridional circulation system attributed to Rossby. Flows patterns are illustrated in Figure 1.

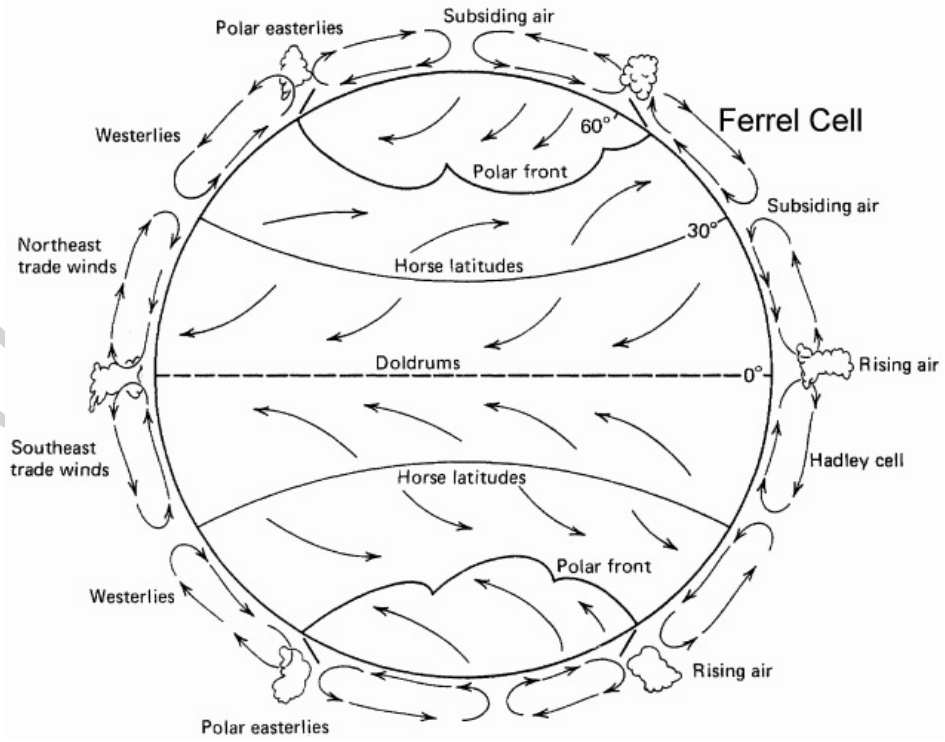

Figure 1. A schematic representation of Rossby's three-cell meridional scheme (adapted from Lutgens and Tarbuck 1986)

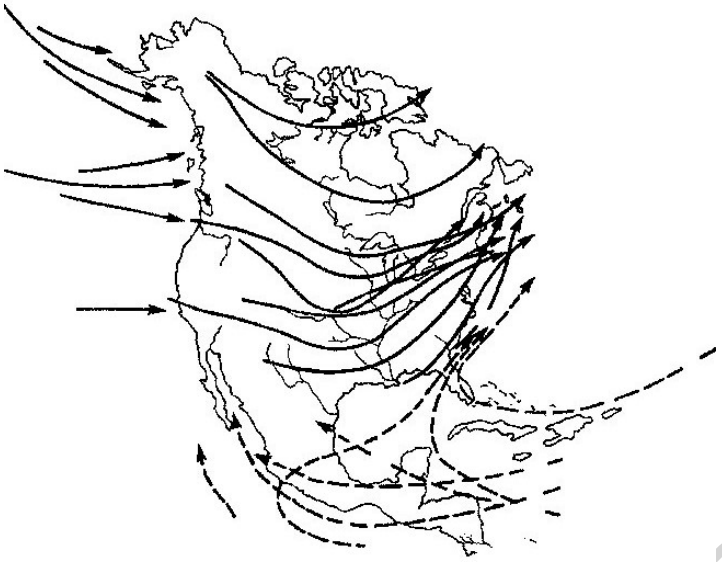

Figure 2. Climatological summary of cyclone tracks over north America (solid lines represent extratropical cyclones and dashed lines denote cyclones, adapted from Irving 1991)

The north-south limits of each of the cells mark the general boundary between the three air masses. However, these boundaries vary seasonally. For example, the Arctic front moves south in northern winter and north in the northern summer. These movements are important factors affecting phenomena such as the Arctic haze and persistent organic pollutant (POPs) influx into the Arctic region as will be described later. In addition, the boundaries between air masses are usually wave-like in nature, with the waves referred to as Rossby waves. These large scale, or planetary, waves, ultimately spawn low pressure systems, or cyclones, which bring clouds and precipitation. In North America, cyclogenesis predominantly occurs in the lee (east) of the Rocky Mountains and along the east coast of North America, and generally propagate to the east and northeast (Figure 2).

Cyclones are also centers of air convergence and result in vertical movements of air. Cyclone development and movement of the cyclone centre obviously modifies airflow and has a fundamental affect on pollutant transport as well as deposition.

Superimposed upon the large-scale air movements, micro-scale dispersal also occurs with time and length scales in the order of hours and kilometers, respectively. The relatively constant wind speeds and direction within the PBL, over these length and time scales, allows simplifying assumptions to be made about the physics of atmospheric dispersal that greatly ease mathematical modeling of air flow and have prompted the development of numerous dispersal models. In the main, these are based on Eulerian or Lagrangian modeling principles and on our understanding of the momentum or pollutant flux relationships within the surface layer and of wind speed variations within the rest of the boundary layer (Arya 1988, Blackadar 1997, Stull 1997).

2.3. Pollutant Deposition

During transport, pollutants may undergo chemical transformation, or depletion from the atmosphere by transfer to the earth's surface by one of two basic mechanisms; wet deposition, wherein pollutants are absorbed by rain, snow or fog and deposited on to the earth's surface (precipitation scavenging), and dry deposition, wherein the pollutants fall or are absorbed at the earth's surface by soil, water or vegetation directly.

(i) Precipitation Scavenging

Particles: As water drops fall through the air they collide with pollutant particles and collect them. The efficiency with which rain drops collect particles is a function of the radii of the rain drops and particles and the fall speed of the rain drop (assuming the pollutant particles have negligible vertical velocity). Integration over the size range of rain drops and pollutant particles, together with data on rainfall rates, yields the total amount of particulate matter that a rain event can collect and transfer to the earth (Seinfeld 1986).

Gases: The capture of particles is assumed to be irreversible; however, gas capture by water drops may be irreversibly or reversibly captured. The scavenging rate of an irreversibly soluble gas is determined by the equilibrium constant between the gas-phase and aqueous phase of the gas, as well as factors such as the droplet size, fall velocity, concentration of the gas and height of droplet fall (Seinfeld 1986). However, the capture rate of a reversibly absorbed gas is determined by the aqueous concentration of the pollutant gas within the droplet as well as the scavenged gas' vapor pressure close to the surface of the drop. Thus, the point of drop saturation must be known or calculated to determine scavenging rate and may vary with drop and environmental conditions (Seinfeld 1986)

(ii) Dry Deposition

Dry deposition of gases or particles is a complex process which is normally conceptualized as being analogous to electrical or heat transfer, and occurring in three consecutive steps:

1. the aerodynamic component, wherein substances are transferred from the atmosphere to the deposition surface, and is largely controlled by atmospheric turbulence and stability,
2. the surface component, wherein substances are transferred through the laminar sub-layer surrounding objects, which is largely controlled by molecular diffusivity, and,
3. the transfer component, wherein substances are absorbed by the surface, and which is controlled by species solubility and/or absorptivity.

Although the laminar sub-layer surrounding objects (plant leaves, soil surface, etc.) may be quite thin (10^{-1} to 10^{-2} cm thick), it is often a crucial step in the entire transfer process. The strength of dry deposition is represented by the deposition velocity, v_d

(units of velocity), which is a constant of proportionality between the atmospheric flux to the surface and the concentration of a particular pollutant at some reference height. The three components of dry deposition are combined in the form of resistances, thus

$$v_d = (r_a + r_s + r_t)^{-1}$$

where r_a is the aerodynamic resistance, r_s is the surface resistance and r_t is the transfer resistance. These components are often derived theoretically or experimentally and readers are referred to various articles (e.g., Brook et al. 1999) and texts (e.g., Seinfeld 1986) for further description. With these concepts firmly in mind it is now useful to describe specific "pollution-based" issues that are of concern and that can be attributed, at least in part, to transboundary movements of pollutants.

3. Smog

3.1. Introduction

Smog is one of most well known pollutants among the general public. It occurs mainly in urban or built-up areas and causes reduced visibility and can make breathing difficult, even for healthy people, and can also increase susceptibility to cardio-respiratory diseases (Burnett et al. 1995). Smog is a complex mix of pollutants including nitrogen oxides (NO_x), sulfur dioxide (SO_2), volatile organic compounds (VOCs), particulate matter (PM) and ozone (O_3), but can also contain many other compounds. Smog issues first came to light during the famous smoke and fog (thus "smog") episodes in London during 1952 for which over 4000 deaths are attributed (Wark et al. 1998), although most smog problems today are produced by photochemical oxidants. Ground-level (as opposed to Stratospheric) ozone is a colorless and highly irritating gas that forms just above the earth's surface. It is called a "secondary" pollutant because it is produced when primary pollutants undergo reactions due to photochemically induced production of free radicals. Two primary pollutants that strongly affect ground-level ozone are NO_x and VOCs. NO_x includes the gases nitric oxide (NO) and nitrogen dioxide (NO_2), and are produced mostly by burning fossil fuels. Ozone not only affects human health, it can damage vegetation and decrease the productivity of some crops, injure flowers and shrubs and may contribute to forest decline (e.g., in Western Europe [Lubkert et al. 1984]) and can also cause collateral damage to manmade materials (Seinfeld 1986). Fine particulates also cause health problems that are described more fully in a later section.

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Biographical Sketches

Philip Fellin

With 29 years in the environmental field, Mr. Fellin has specific expertise in:

-- measuring air pollutants

-- indoor air quality and occupational hygiene measurements

-- developing analytical methods, instrumentation and methods for sampling airborne compounds

-- designing studies to investigate chemical and physical processes in the atmosphere such as the long range transport of airborne pollutants (acid rain), urban air pollution (smog), fugitive emissions from industrial facilities, and

-- measuring personal exposures in occupational, indoor and transportation environments

In addition, he has been involved in the measurement of airborne toxic compounds in both populated and remote areas such as the Arctic, executed environmental impact assessments for new industrial facilities and delivered training programs on air pollution measurement and assessment in Canada and internationally.

Franco DiGiovanni has had 10 years dispersal modeling experience and is presently Air Quality and Dispersal Modeler for Airzone One Ltd. He has particular expertise in particulate matter dispersal modeling and measurement. He is also experienced in field program execution aimed at model verification. In addition, he has responsibilities in the bioaerosol (ambient and indoor) and indoor air quality sectors, and in emissions estimation and regulatory compliance.