

Long-Range Transport of Chemicals in the Environment

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Foreword

The Monitoring & Environmental Chemistry Working group (MECW) is a Euro Chlor science group which represents the European chlor-alkali industry. The main objectives of the group are to identify both natural and anthropogenic sources of chlorinated substances, study their fate, gather information on the mechanisms of formation and degradation in the environment and achieve a better knowledge of their environmental persistence. The MECW often uses external specialists to assist in developing reports that review the latest knowledge of the different aspects mentioned. The principal investigator collects information from the scientific literature and available data regarding natural or anthropogenic emissions in the environment to provide a comprehensive overview.

Dr James Franklin, a physical chemist by training, founded his own environmental consulting company in 2004. He has a particular interest in atmospheric chemistry. Prior to setting up his own business, Dr Franklin worked in the central research department of Solvay (Brussels, Belgium) for more than 33 years. During this time he worked mainly on the development of production processes for organic chlorine and fluorine compounds as well as on their applications and how they impact the environment.

Long-range transport (LRT) of chemicals is a key issue that continues to attract the attention of scientists, NGOs, environmental journalists and general public. The reason for such wide-spread interest is that the environmental impact of chemical use not only remains at their place of origin, but can be carried by air currents, surface water or migrating animals to regions around the globe.

This report examines the scientific literature on experimental observations demonstrating how natural and man-made substances are transported to locations remote from their point of release. It also looks at the timescales involved in atmospheric long-range transport including the concepts of "global fractionation" and the "grasshopper effect". Dr Franklin reviews the mathematical models used to simulate atmospheric distribution, degradation and transport of chemicals. He also looks at the models used to compare LRT potential of different substances and determine the sources of pollutants arriving at a given receptor location or how the receptors are affected by a given source. In addition to the above, the report covers the international regulatory agreements set up to reduce the trans-boundary LRT of pollutants and the criteria adopted in different conventions for expressing the propensity of a substance to undergo LRT.

The potential impacts of LRT on human health and ecosystems are mentioned, but not analysed in any depth. No assessment of the risks associated with the LRT of specific substances is provided.

Summary

Long-range transport (LRT) of chemicals is a key issue being addressed by politicians at international level due to the far-reaching consequences of the phenomenon. As a result of the mobile nature of the Earth's atmosphere, every region of the globe is affected by air pollutants. Often, they travel thousands of miles from where they originate. As well as air, water and migratory animals also aid this process.

The time it takes for a substance to move from one area of the world to another depends on the substance and atmospheric conditions. Intercontinental atmospheric transport generally occurs over three to 30 days. Pollutants that occur in the troposphere will typically travel further and faster than those that are advected horizontally in the atmospheric boundary layer. On the other hand, dissolved species transported by ocean currents from northern mid-latitudes to the Arctic can take several years.

Volatile pollutants with atmospheric lifetimes of years or longer experience no impediment to LRT once they are emitted to air. Whatever the location and timing of release, their concentrations become practically uniform throughout the atmosphere, so the emission of a given amount anywhere on Earth makes the same contribution to global pollution. For air pollutants with lifetimes of several months, concentrations become uniform within the hemisphere of emission, but not in the opposite hemisphere. Crossing the equator takes about a year. For shorter-lived pollutants with atmospheric lifetimes of days or weeks, the tendency for the substance to undergo LRT depends on its physico-chemical properties as well as how it entered the environment, the location and time of its emission and prevailing meteorological conditions.

Trans-boundary movement applies to man-made substances such as pesticides and lead, as well as part man-made - part natural substances such as carbon monoxide and nitric oxide. Similarly, purely natural substances, including desert sand and sea salt, are subject to such environmental displacement.

LRT is not in itself a concern, but only a trigger for potential concern. In many cases, however, deposition followed by uptake into living organisms and possible accumulation through the food chain, is a prerequisite for damage to the environment and human health. Each chemical should be evaluated on a case-by-case basis, with the LRT potential assessed in conjunction with properties such as persistence, bioaccumulation, toxicity and the capacity to affect the atmosphere.

1 Introduction: What is meant by Long-Range Transport of chemicals and why is it of interest?

In this chapter, a brief introduction is given to the concept of Long-Range Transport (LRT) of chemicals in the environment and its consequences. All the aspects introduced in this section will be developed in greater depth in the following chapters of this dossier.

The “Transboundary” nature of air pollution

On account of the mobile nature of the Earth’s atmosphere, all regions unwillingly – and in some cases unwittingly – receive air pollutants not only from their close neighbours but also from further afield. In turn, the substances these regions themselves release to air can be transported to other territories.

This phenomenon has become particularly acute with the rise of industrialisation, but long-range atmospheric transport of man-made pollutants is now known to have existed, to some extent, for millennia.

Transport through media other than air

Air is the medium most often considered responsible for the LRT of chemicals in the environment on account of its ubiquitous nature and its facility of movement. However, other mobile media may also contribute to such transport, depending on the physico-chemical properties and point of release of the substances concerned.

The most noteworthy of these other media are water (oceans, rivers and drifting ice) and, to a lesser extent, migratory animals. For the sake of completeness, one should also include anthropogenic transport, for commercial or other purposes, which can contribute to disseminating chemicals in the environment far from where they are produced or used.

The global nature of Long-Range Transport

The potential of a chemical for LRT, sometimes referred to as its “characteristic travel distance” or “spatial range”, depends not only on its intrinsic physico-chemical properties, but also on its mode and point of release to the environment. The prevailing meteorology or ocean currents are very important.

LRT is generally taken to mean transport over distances of at least a few hundred kilometres, i.e. far enough for the substance to cross the borders of smaller nations. However, intercontinental transport, over thousands or even tens of thousands of kilometres, has also become the focus of particular concern, especially in connection with the pollution of the erstwhile pristine regions within the polar circles.

Well-established examples of LRT will be provided in this dossier, for a wide range of anthropogenic and natural substances. The mechanisms and preferential pathways will be outlined, together with the models used to simulate transport and the methodologies developed for assessing LRT potential and investigating source-receptor relationships.

Consequences of Long-Range Transport

LRT becomes relevant only when the chemical transported has some adverse impact on human health or on ecosystems far from its source.

Certain volatile compounds may undergo LRT without being deposited to the Earth’s surface, but may nevertheless exert a deleterious influence on the atmosphere itself (through ozone depletion, contribution to greenhouse warming, photochemical smog formation, etc.) or on human health by inhalation.

Other substances may lead to harmful effects only when they are deposited from the atmosphere to land or water bodies.

It should therefore be emphasised that LRT is not, in itself, a concern, but only a trigger for potential concern. When evaluating a chemical's hazard potential, LRT needs to be assessed in conjunction with adverse properties, such as persistence, bioaccumulation, toxicity and the capacity to affect the atmosphere.

The ethical dimension of Long-Range Transport and the enacting of international regulatory agreements

The perceived injustice of passively undergoing the consequences of some other nation's pollutants, at the whim of air or water currents, has led to the enacting of a number of international agreements aimed at mitigating the release and impact of persistent, bioaccumulative and toxic pollutants. These agreements will be discussed, together with the "criteria" they adopt for characterising the propensity of a substance to undergo LRT.

2 Selected examples of observational evidence for atmospheric Long-Range Transport

All regions of the globe are affected to a greater or lesser extent by the atmospheric LRT of chemicals. The scientific literature on the subject is vast and the present dossier is not intended to give a comprehensive coverage of it. The aim is merely to illustrate the phenomenon with a limited number of well-documented examples for a broad range of substances, not all of which are man-made.

Some of the substances discussed are deposited to ground level following atmospheric LRT and may thus exert harmful effects in the biosphere. Others are too volatile to undergo significant deposition, but may lead to an undesirable impact through their presence in the atmosphere (contribution to ozone depletion, global warming, photochemical smog, etc.). Furthermore, their degradation products may be deposited.

2.1 Substances of natural origin

2.1.1 Sand, dust and ash

For many citizens in their everyday life, the most tangible evidence for atmospheric LRT is provided when wind-borne sand originating from a faraway desert casts a reddish-yellow blanket on their newly washed automobiles, or when it blemishes the surface of freshly fallen snow.

The LRT of desert sand, or of clayey "loess" dust, has been observed to occur episodically or according to a seasonal cycle and to follow a number of trajectories worldwide (Rea, 1994; Simpson *et al*, 2003a; Husar, 2004). Some prime examples of such transport, occurring over time-scales of days or weeks, are the following:

- from the Sahara/Sahel region westwards to the Caribbean and the central part of the American continent or from North Africa to the Mediterranean area and Europe, as far as the United Kingdom and the Baltic Sea, or even eastwards to Asia (Moulin *et al*, 1997; Herman *et al*, 1997; Alpert and Ganor, 2001; Griffin *et al*, 2001a; DeMott *et al*, 2003; Ansmann *et al*, 2003; Grousset *et al*, 2003; Piketh and Walton, 2004; Rogora *et al*, 2004; Collaud Coen *et al*, 2004; Aymoz *et al*, 2004; Sodemann *et al*, 2005; Tanaka *et al*, 2005; Barkan *et al*, 2005; Chiapello *et al*, 2005);
- from the Gobi Desert and other areas in China and Mongolia, eastwards across the Pacific Ocean and North America (Wilkening *et al*, 2000; Husar *et al*, 2001; Sassen, 2002; Takemura *et al*, 2002; Jaffe *et al*, 2003a; Gong *et al*, 2003; DeBell *et al*, 2004; Perry *et al*, 2004), occasionally reaching Greenland and Europe, after covering a distance of more than twenty thousand kilometres (Biscaye *et al*, 2000; Grousset *et al*, 2003);
- from southern Africa to Australia, and from Australia to New Zealand (Sturman *et al*, 1997; Marx *et al*, 2005).

Dust storms occurring in Asia have been shown to be responsible for nucleation of ice clouds over Alaska at temperatures far warmer than those expected for normal cirrus cloud formation (Sassen, 2005).

Surface aerosol concentration enhancements at northern mid-latitudes arising from intercontinental transport of dust have been reviewed by Holloway *et al* (2003). With the spread of desertification, LRT of dust can be expected to become an increasing concern.

Volcanic ash can undergo LRT in a similar manner to sand and dust (Rea, 1994; Simpson *et al*, 2003a).

Satellite imagery provides a vivid confirmation of the intercontinental transport of dust and other particles, as well as of certain gases (e.g. Hsu *et al*, 1999; Engel-Cox *et al*, 2001; Wooster and Strub, 2002; Damoah *et al*, 2004).

2.1.2 Sea salt

Another abundant natural mineral substance undergoing atmospheric LRT is sea salt (Gustafsson and Franzén, 2000; Fischer, 2001; Gong *et al*, 2002). However, LRT is efficient only for a fraction of the aerosol particles, namely the sub-micron sized ones, which remain aloft longer than their larger counterparts, unless the latter are lifted into the upper troposphere by forceful convection such as can occur in the tropics (Ikegami *et al*, 1994).

2.1.3 Natural radionuclides

The naturally-occurring radioisotope ^{210}Pb has been widely used for investigations of transport and removal processes in the lower troposphere (Rehfeld and Heimann, 1995; Dueñas *et al*, 2003; Lee *et al*, 2004). It is formed by the decay of ^{222}Rn emanating from the Earth's crust. This radon isotope has a relatively short half-life (3.8 d) and is therefore concentrated mainly in the continental planetary boundary layer. On the other hand, the LRT of ^{210}Pb , with a decay half-life of 22 y, is limited only by physical removal processes. Transport of this isotope to the polar regions has been demonstrated (Lambert *et al*, 1990).

2.1.4 Volatile organic compounds

For want of a better descriptor, the term “volatile organic compound” (VOC) will be used in this dossier to denote any organic species that partitions preferentially to the atmosphere and that has a lifetime of at least a few days, so that LRT becomes possible. Note that this usage is somewhat different from the terminology adopted in connection with the formation of tropospheric ozone. In that case, “VOC” is reserved for the particularly reactive organic gases (with lifetimes of hours or days) that are essential ingredients in local “photochemical smog” production.

Among the volatile organic compounds of almost exclusively natural origin are certain halogenated aliphatic species: CH_2Br_2 (approximate lifetime 0.3 y), CH_2BrCl (0.4 y), CHBr_3 (4 weeks), CHBr_2Cl (0.2 y), CHBrCl_2 (0.2 y), CH_3I (1 week), $\text{C}_2\text{H}_5\text{I}$ (4 d) and CH_2ICl (a few hours) (Solomon *et al*, 1994; Singh and Fabian, 1999; Baker *et al*, 2001; Ballschmiter, 2003; Keppler *et al*, 2003; Harper and Hamilton, 2003; Quack and Wallace, 2003; Quack *et al*, 2004; Chuck *et al*, 2005; Zhou *et al*, 2005).

On account of their low water solubility and high volatility, none of these organic halogen compounds will be removed significantly from the atmosphere by wet or dry deposition. The ones with lifetimes of months or more are long-lived enough to become fairly well distributed throughout the hemisphere in which they are emitted (WMO, 2002; Thompson *et al*, 2004). Following atmospheric LRT, only their degradation products (i.e. mainly hydrohalic acids) will reach the biosphere.

2.1.5 Biota

Pathogenic microbes can be transported over long distances through the atmosphere and hence potentially impact ecosystems and human health far from their source region. For example, the levels of various cultivable airborne plant and human pathogens were shown to increase sharply in the Caribbean following African dust events (Griffin *et al*, 2001a). Furthermore, it has been observed that viable fungal spores can be transported not only with mineral dust, but also with smoke from biomass fires (Mims and Mims, 2004). Australian pollens and other organisms are regularly transported by wind to New Zealand (Sturman *et al*, 1997). Muñoz *et al* (2004) provide evidence in support of the hypothesis that the close affinities of the floras of the continents and islands in the southern hemisphere are the consequence of LRT of airborne spores and seeds.

Griffin *et al* (2001b) provide a fascinating review of the LRT of airborne pathogens, including those responsible for meningococcal meningitis, potato blight, foot-and-mouth disease and a whole host of other plant and animal afflictions.

Atmospheric monitoring and modelling studies have been used to detect illegal cultivation of marijuana, by combining pollen measurements in the ambient air in Spain with the back-analysis of trajectories originating in North Africa (Cabezudo *et al*, 1997).

Even macro-scale living organisms can be subject to a form of “assisted” atmospheric LRT. Thus, the displacements of swarms of African desert locusts - westwards across the

Atlantic, or northwards to Europe - are made possible not only by the insects' intrinsic flight capabilities, but also by the prevailing winds (Rosenburg and Burt, 2001).

2.2 Substances Having both Natural and Anthropogenic Sources

This section provides examples of the LRT of a range of compounds that have significant natural sources as well as originating from human activities.

2.2.1 Major greenhouse gases: Carbon dioxide, methane and nitrous oxide

Atmospheric emissions of carbon dioxide are mainly of natural origin and sources and sinks of CO₂ were in equilibrium until the beginning of the industrial era. However, since then, combustion of fossil fuels and other human-related activities have led to additional emissions of CO₂, disturbing the balance. The resulting rise in concentrations of CO₂ and the "greenhouse effect" that they are believed to cause have led to numerous scientific studies and regulatory initiatives that are beyond the scope of this dossier. However, it is appropriate to point out that the atmospheric persistence of CO₂ is long enough (i.e. centuries) for this gas to become well mixed throughout the atmosphere, so emissions anywhere on the planet will contribute to the global background of CO₂ and the climate effects ascribed to it.

The same conclusion is valid for two other important greenhouse gases that are of partly natural and partly anthropogenic origin: methane (lifetime 12 years) and nitrous oxide (lifetime 114 years) (Conway *et al*, 2004).

2.2.2 Carbonaceous particles

Together with mineral dust and water-soluble inorganic salts, carbonaceous particles are a major component of the overall aerosol burden of the lower atmosphere. They are formed and emitted, together with a broad variety of volatile compounds, during the incomplete combustion of biomass, fossil fuels and other organic substances. Since biomass burning includes both forest and grassland wildfires, agricultural burning, and domestic fires for heating and cooking purposes, carbonaceous aerosols clearly arise from both natural and anthropogenic sources (Cooke and Wilson, 1996; Liousse *et al*, 1996; Jacobson *et al*, 2000).

From a historical perspective, it is interesting to note that in 18th century England, shepherds high up on the moors already observed "moorgroime", a black deposit on the wool of sheep that originated from coal burning in the cities. In the following century, there was some evidence for "coal dust" from Britain reaching Norway (Brimblecombe, 2001).

Today, fossil-fuel combustion in the main industrialised regions of North America, Europe and Asia makes a major contribution throughout the year to carbonaceous particle emissions. In addition, huge vegetation fires break out regularly in both the tropics (Africa, South America and South East Asia) and in the boreal and mid-latitude regions (Canada, the United States, Russia, Scandinavia and the Mediterranean area). Furthermore, agricultural biomass burning occurs particularly in the tropics during the dry season (Crutzen and Andreae, 1990).

In addition to the "primary" emissions of solid particles, "secondary" organic aerosols can be formed in the atmosphere by reactions involving gaseous natural or anthropogenic precursor compounds, especially the biogenic terpenes (Jacobson *et al*, 2000; Kanakidou *et al*, 2000).

While the chemical composition and physical properties of the carbonaceous particles formed are highly complex, variable and only partially characterised as yet, it is recognised that such aerosols can affect public health, weather, climate, visibility, etc.. Understanding the atmospheric LRT of these particles is therefore essential for assessing the impact they may exert far from their source regions.

In addition to elemental carbon, such particles contain a range of organic substances, some of which may be harmful (e.g. the carcinogenic polycyclic aromatic hydrocarbons). Furthermore, the particles can also act as "sponges", taking up low-volatility compounds from the gaseous phase of the atmosphere and hence determining the LRT patterns of the latter.

Certain specific biomass thermal degradation products present in the particles act as tracers or “biomarkers”, enabling the nature of the source to be identified and hence providing evidence for the occurrence of LRT (Simoneit and Elias, 2001; Simoneit, 2002).

The emissions and transport of carbonaceous aerosols from natural and anthropogenic sources have been simulated on a global scale using a number of three-dimensional models (Cooke and Wilson, 1996; Liousse *et al*, 1996; Kanakidou *et al*, 2000; Koch, 2001; Chung and Seinfeld, 2002; Damoah *et al*, 2004). Convective processes due to fires can convey the combustion products and the substances derived from them into the free troposphere – and even the lower stratosphere (Fromm *et al*, 2000) – while large-scale atmospheric circulation distributes them to locations far from their source regions (Crutzen and Andreae, 1990; Spichtinger-Rakowsky and Forster, 2004; Damoah *et al*, 2004).

To some extent, carbonaceous particles may follow the atmospheric transport pathways described above for desert sand and dust. However, since different source regions are involved, other trajectories will also come into play.

The pollution plumes from Canadian forest fires have been shown to affect the eastern and southeastern United States (Wotawa and Trainer, 2000; Sapkota *et al*, 2005), while those from both Canadian and U.S. fires reach continental Europe within about 1 week, after passing over Greenland and the Atlantic (Hsu *et al*, 1999; Forster *et al*, 2001; Fiebig *et al*, 2002).

Smoke from a severe forest fire in Russia in 2003 was shown to circumnavigate the globe in 17 days (Damoah *et al*, 2004). One plume travelled eastwards to Alaska, Canada, and Scandinavia, and then on to Eastern Europe, where it merged with smoke advected westwards from the same fire. A westward path to Sweden and Germany was also taken by a plume from a peat fire near Moscow in 2002 (Spichtinger-Rakowsky and Forster, 2004). Smoke from Russian forest fires has also been shown to affect the Korean Peninsula (Lee *et al*, 2005).

Smoke arriving in Greenland could be attributed not only to North American sources, but also to fires occurring in Russia and China (Hsu *et al*, 1999). Observations on black carbon in the Arctic have been reviewed by Iversen (1996) and Bottenheim *et al* (2004). In winter and spring, industrial regions in Eurasia are the major sources of carbonaceous aerosols reaching Barrow, Alaska, with some contribution from western North America and Western Europe (Polissar *et al*, 2001).

Particles arising from combustion sources in Asia were demonstrated to reach the U.S. west coast (Jaffe *et al*, 2001; Bertschi and Jaffe, 2005). Martin *et al* (2002) showed that not only could polluted air parcels from Asia cross the Pacific to North America, but in certain cases they might pick up further contaminants there before looping back over the Pacific towards Indonesia. Furthermore, anthropogenic emissions from Europe and biomass burning emissions from Africa make major contributions to the Asian outflow over the Pacific (Bey *et al*, 2001).

Depending on their point of origin, emissions from biomass burning in southern Africa can be carried westwards over the Atlantic or eastwards over the Indian Ocean, in some cases reaching Australia (Herman *et al*, 1997; Sturman *et al*, 1997; Piketh and Walton, 2004) or the Pacific islands of Samoa and Fiji (Thompson, 2004).

Pollution from biomass burning in tropical or sub-tropical South America can be exported either eastwards over the Atlantic or westwards over the Pacific (Schultz and Bey, 2004).

Vegetation fires raging on the islands of Sumatra and Borneo in 1997-1998 cast a blanket of smog over the region, including the Malaysian Peninsula, exposing more than 20 million people to potentially dangerous levels of air pollution for many months (Heil and Goldammer, 2001; Wooster and Strub, 2002; Langmann and Heil, 2004). The “brown cloud” arising from biomass and fossil fuel burning is a regular feature over broad areas of Asia (ASEAN, 2001; UNEP-C⁴, 2002).

Fall-out from Kuwaiti oil-field fires in 1991 was traced as far as Hawaii (Lowenthal *et al*, 1993). Similarly, elemental carbon from oil fields burning in Iraq in March 2003 was deposited in Japan a few days later (Tazaki *et al*, 2004).

In the same way as for dust particles, the lifetimes of carbonaceous aerosols, up to a week or so (unless they are lofted into the stratosphere), are far too short for them to cross from one hemisphere to the other through the “intertropical convergence zone”. This process involves an exchange time of about one year (Ballschmiter, 1992).

2.2.3 Other combustion by-products and related oxidants: CO, NO, O₃, etc.

The various combustion processes mentioned above in connection with the formation of carbonaceous aerosols lead to a wide range of by-products. This includes carbon monoxide, reactive organic substances (more commonly categorised as VOCs and nitric oxide, all of which are precursors in the formation of tropospheric ozone and the related oxidant peroxyacetyl nitrate (PAN).

Ozone formation is often considered to be a problem associated with large cities or industrial areas in summer. However, since combustion by-products are subject to LRT, they contribute to rising ozone levels in background air far from populated regions, a phenomenon sometimes known as “global smog”, for which a major driver is biomass burning (Fishman *et al*, 1996; Lelieveld *et al*, 2004).

Apart from any specific environmental or health impact the combustion by-products may exert by themselves, a related concern is therefore the ozone they are capable of generating far from their point of emission.

Furthermore, ozone present in the free troposphere – that is above the planetary boundary layer which extends to about 1 km altitude – has a lifetime of a few weeks (Thompson, 2004). This is long enough for it to undergo LRT itself, in addition to any transport of its precursors, such as NO₂ whose lifetime is only about two days in the boundary layer, but up to a week in the middle and upper troposphere (Wenig *et al*, 2003; Schaub *et al*, 2005), and CO which has a lifetime of months (Seinfeld and Pandis, 1998, pp. 85-86).

The global atmospheric pathways travelled by combustion by-products are similar to those described above for the carbonaceous aerosols having the same origin. They are however not necessarily identical, mainly on account of the species-dependent chemical and physical removal processes, which determine the lifetimes of the individual substances.

Nevertheless, to avoid excessive repetition, these transport pathways are not described in detail. Further information on LRT of ozone and its precursors can be found in the following recent papers and the references cited therein: Bertschi and Jaffe (2005); Bey *et al* (2001); Bottenheim *et al* (2004); Cooper and Parrish (2004); Cooper *et al* (2004a, 2004b); Creilson *et al* (2003); de Gouw *et al* (2004); Duncan and Bey (2004); Formenti *et al* (2002); Forster *et al* (2001); Goldstein *et al* (2004); Heald *et al* (2003); Holzer *et al*, 2003; Hudman *et al*, 2004; Huntrieser and Schlager (2004); Huntrieser *et al* (2005); Jacob *et al* (1999, 2003); Jaffe *et al* (2001, 2003b, 2004); Kato *et al*, 2002; Langmann and Heil (2004); Langmann *et al* (2003); Lawrence (2004); Li *et al* (2002); Liu *et al* (2002, 2005); Martin *et al* (2002); Naja *et al* (2003); Parrish *et al* (2004); Penkett *et al* (2004); Piketh and Walton (2004); Pochanart *et al* (2001, 2004); Price *et al* (2004); Schaub *et al* (2005); Schultz and Bey (2004); Stohl and Trickl (1999); Spichtinger *et al*, 2001; Spichtinger-Rakowsky and Forster (2004); Stohl *et al* (2002a); Thompson (2004); Trickl *et al* (2003); UNECE (2003); Wenig *et al* (2003); Wild and Akimoto (2001); Wilkening (2001); Wilkening *et al* (2000); Wotawa and Trainer (2000); Wotawa *et al* (2001); Yienger *et al* (2000).

It is interesting to note that, according to modelling studies, there is a significant contribution both of North American anthropogenic emissions to surface levels of ozone in Europe and of European emissions to levels of ozone in North America, at least under certain meteorological conditions (Li *et al*, 2002; Guerova *et al*, 2005; Auvray and Bey, 2005). Asia is affected by emissions from both Europe and North America (Liu *et al*, 2002). Roelofs *et al* (2003) demonstrated that ozone levels in the eastern Mediterranean area were significantly influenced by emissions from Southeast Asia, North America and Europe. Wild and Akimoto (2001) suggested that European fossil fuel emissions had a lower potential than Asian or North American emissions to add to ozone levels in the middle and

upper troposphere. Lelieveld *et al* (2002) showed that in summer the middle troposphere of the Mediterranean area receives a westerly inflow of CO and other pollutants from Asia and North America, while the upper troposphere is influenced by Asian pollution coming from the east.

Ozone pollution is clearly shifting from a purely local concern to a global issue, since LRT of O₃ and its precursors is now recognised to contribute significantly to regional budgets (Creilson *et al*, 2003; Holloway *et al*, 2003; Chatfield *et al*, 2004; Schultz and Bey, 2004; Sauvage *et al*, 2005).

2.2.4 Sulphur dioxide and sulphate aerosols

Atmospheric emissions of sulphur dioxide result mainly from the combustion of sulphur-containing coal and other fossil fuels, but the contributions from volcanoes and the biogenic precursor dimethyl sulphide are non-negligible (Benkovitz *et al*, 2004). Gas-phase and in-cloud oxidation of SO₂ leads to sulphuric acid. Together with nitric acid arising from combustion-derived NO and other anthropogenic and natural sources of nitrogen oxides, sulphuric acid is one of the main components of “acid rain” (Bouwman *et al*, 2002).

Air pollution by long-range transported SO₂ is not a recent problem. Seinfeld and Pandis (1998, p. 1030) cite an incident occurring three centuries ago, when SO₂ emitted from “industry” in the United Kingdom travelled far downwind and remained at a high enough concentration to cause bleaching of dyed cloth in France.

Despite wet scavenging of SO₂ as sulphate after oxidation in the gaseous and aqueous phases, intercontinental transport of SO₂/sulphate undoubtedly occurs, as demonstrated by the following examples:

- instances of trans-Pacific transport of SO₂/sulphate from Asia to the Californian coast have been reported (Brock *et al*, 2004);
- sulphate deposition in the Canary Islands has been shown to be due mainly to North American sources under certain meteorological conditions (Benkovitz *et al*, 2003);
- 10 % of wet deposition of sulphur over western Europe was calculated to arise from North American emissions (Tarrasón and Iversen, 1992);
- the boundary layer “haze” that afflicts the Arctic, particularly in winter and spring, is made up mainly of sulphate aerosols arising from SO₂ emissions in Europe and the former Soviet Union, while sources further from the Arctic continent also contribute significantly to pollution by sulphur species in the free troposphere (Barrie *et al*, 1989; Iversen, 1989, 1996; Bottenheim *et al*, 2004; Heidam *et al*, 2004).

Furthermore, transport between neighbouring countries on a regional scale is still a major concern, despite considerable reductions in SO₂ emissions in the developed nations over the last few decades.

For example, rapid economic development in China, coupled with the use of coal as a major source of energy, is considered to make a significant contribution to sulphuric acid deposition in other Asian countries and even the central northern Pacific (Carmichael *et al*, 2002; Tu *et al*, 2004), although quantitative modelling is subject to uncertainties, on account of the sensitivity to wet removal parameterisation.

Many of the smaller sulphur-emitting countries in Asia and the Indian sub-continent receive more sulphur deposition than they emit (Arndt *et al*, 1998).

South-East Asia (Indonesia, Malaysia, The Philippines and Singapore) is estimated to receive a quarter of its sulphur deposition from shipping activities (Guttikunda *et al*, 2001).

Sulphur dioxide emissions from southern Africa are believed to reach Amsterdam Island, thousands of kilometres away, while those from Australia may well affect New Zealand (Sturman *et al*, 1997).

Pollution of the Aegean Sea and eastern Mediterranean area by sulphate aerosols and other substances originating from emissions in continental Europe has been discussed by Wanger *et al* (2000) and Formenti *et al* (2002).

Benkovitz *et al* (2001) have carried out modelling studies to show how the atmospheric transport and loading of SO₂ and sulphate are influenced by weather patterns over North America, the North Atlantic and Europe. For instance, under certain meteorological conditions, it was shown that transport of SO₂/sulphate from Europe to Iceland was likely.

Volcanoes can inject SO₂ and other substances directly into the stratosphere. Thus the 1991 eruption of Mount Pinatubo in the tropics (15°N) lofted tens of millions of tons of SO₂ and sulphate up to an altitude culminating close to 40 km. The resulting aerosol cloud, travelling westwards, straddled the equator and encircled the Earth in 21 days (Bluth *et al*, 1992; Guo *et al*, 2004). Over a period of months, the sulphate aerosol spread towards the poles and ultimately contributed significantly to ozone depletion (Solomon, 1999) and climate cooling (Robock, 2002) on a global scale.

2.2.5 Mercury

Mercury is a volatile metal. The atmosphere plays a major role in the mobilisation and geographical redistribution of this element, for which both human-related and natural emissions are significant (Fitzgerald *et al*, 1998; Lin and Pekhonen, 1999; Bergan *et al*, 1999; UNEP, 2002b; Ilyin *et al*, 2004).

Anthropogenic releases of mercury to air arise mainly from fossil fuel combustion, especially coal burning in Asia. The production of metals and cement, along with waste incineration, also make non-negligible contributions to emissions (Pacyna and Pacyna, 2002).

Although recent studies suggest that elemental mercury's atmospheric lifetime is only a few months, instead of up to two years as believed previously (Renner, 2004), this persistence is still sufficient for mercury to become broadly distributed at least throughout the hemisphere in which it is emitted.

The main concern with atmospheric mercury is its oxidation to "reactive gaseous species" that are readily scavenged to ground level by settling particles and rain. These compounds can then undergo biologically mediated methylation to the bioavailable and toxic monomethylmercury, which can accumulate in terrestrial and aquatic food chains (Schroeder and Munthe, 1998; Ullrich *et al*, 2001; UNEP, 2002b).

Prime evidence for atmospheric LRT of mercury is provided by long-term Hg concentration profiles in cores taken from lake sediments and peat bogs far from local sources. These studies show rising levels over the last century (Fitzgerald *et al*, 1998; Bottenheim *et al*, 2004; Skov *et al*, 2004a; Perry *et al*, 2005; Steinnes and Sjøbakk, 2005).

Pollution of the Arctic, which itself has no significant mercury emissions, by Hg released to air in populated areas at lower latitudes has been the subject of numerous investigations and reviews. Sources in both Eurasia and North America have been implicated (Macdonald *et al*, 2000a; Cheng and Schroeder, 2000; Lin *et al*, 2001; AMAP, 1998, 2002; Schroeder *et al*, 2002; Bottenheim *et al*, 2004). Modelling studies on transport to the Arctic have been performed recently (Heidam *et al*, 2004; Christensen *et al*, 2004), based on a global inventory of emissions of mercury from anthropogenic sources (Pacyna and Pacyna, 2002). "Mercury depletion events", occurring during the Arctic (and Antarctic) spring after the polar sunrise and mediated by halogen species, lead to rapid oxidation of elemental mercury and the resulting efficient deposition of oxidised forms (Lindberg *et al*, 2002; Ebinghaus *et al*, 2002; Bottenheim *et al*, 2004; Heidam *et al*, 2004; Skov *et al*, 2004a, 2004b; Fitzgerald *et al*, 2005; Douglas *et al*, 2005).

In Antarctica, during the austral summer, reactive gaseous mercury levels have been found to be as high as in some industrial environments (Sprovieri *et al*, 2002).

A global modelling study (Dastoor and Larocque, 2004) indicates frequent episodes of mercury transport from Europe to North America via the Arctic, in winter. On the other hand, in summer, mercury can be transported from North America to the northern Atlantic Ocean and Europe.

Evidence has been provided for the transport of mercury across the northern Pacific from Asia to California (Steding and Flegal, 2002, 2003; Jaffe *et al*, 2003c, 2005). Modelling

studies (Seigneur *et al*, 2004) indicate that a significant fraction (5-36 %) of overall mercury deposition at various locations in the continental United States is due to Asian emissions.

The results of an intercomparison of numerical models for atmospheric LRT of mercury have been presented by Ryaboshapko *et al* (2004).

Ilyin *et al* (2004) document the greatly decreasing deposition in European countries of mercury arising from transboundary transport, over the timeframe 1990-2000.

2.2.6 Volatile organic compounds

As discussed above, a number of "volatile organic compounds" (defined for the purposes of this dossier as being any organic species that partitions preferentially to the atmosphere and that has a lifetime of at least a few days) are of overwhelmingly natural origin. Many others have natural and anthropogenic sources that are both significant. One such example is methane, already discussed above as a major greenhouse gas. But a whole range of other organic species also fall into this category, including ethane, propane, butane, benzene, acetone, methanol, etc. (Seinfeld and Pandis, 1998). Indeed, lower aliphatic hydrocarbons ascribed to natural gas emissions in the Gulf of Mexico were encountered over the Mediterranean area (Mühle *et al*, 2002), while a signal from industrial propane emissions in northern Africa was identified out over the Atlantic Ocean (Gros *et al*, 2004).

Aliphatic halogen compounds having significant sources of both natural and anthropogenic origin are CH₃Cl (lifetime 1.3 y), CHCl₃ (0.4 y) and CH₃Br (0.7 y) (Khalil, 1999; Khalil *et al*, 1999; Laturus *et al*, 2002; Ballschmiter, 2003; Harper and Hamilton, 2003; McCulloch, 2003). These compounds degrade in the atmosphere and only their breakdown products (essentially HCl and HBr) reach the biosphere.

2.3 Substances Primarily of Anthropogenic Origin

2.3.1 Anthropogenic radionuclides

For the general public, one of the most vivid, albeit tragic, demonstrations of the reality of atmospheric LRT was experienced after the explosion occurring in the nuclear reactor at Chernobyl, Ukraine, in April 1986. Following this largest reported accidental release of radioactive material, the initial pollution plume dispersed to the northwest and reached Finland and Sweden. However, broad areas of Europe were soon blanketed by a radioactive cloud (Anspaugh *et al*, 1988; Pöllänen *et al*, 1997; Brandt *et al*, 2002; UNEP, 2002a; Robertson, 2004). In Finland, the first temporary increases in radioactivity were observed 1-2 days after the initial explosion (Jylhä, 2000a, 2000b). Fission products from Chernobyl, such as ¹³⁷Cs and ¹³⁴Cs, were traced as far away as Japan (Aoyama, 1988; Kasamatsu and Inatomi, 1998). Lauritzen and Mikkelsen (1999) compared measured and modelled ¹³⁷Cs activities arising from Chernobyl up to distances of 9000 km from the source. A significant increase in radioactivity in the indigenous people of Arctic Norway, Sweden and Russia was observed in the few years immediately following the accident (UNEP, 2002a).

Another case of accidental atmospheric release of ¹³⁷Cs occurred at a steel mill in Algeciras, Spain, in May 1998. This radio-isotope was subsequently detected in air across broad areas of Europe (Pobanz *et al*, 1999; Bysiek *et al*, 2001; Robertson, 2004).

Radionuclides, in particular ⁹⁰Sr, were also dispersed on a global scale by the atmospheric nuclear weapons testing in the 1950s and 1960s (Rehfeld and Heiman, 1995; Dueñas *et al*, 2003).

2.3.2 Lead

Anthropogenic sources of lead include its use as a petrol/gasoline additive (now phased out in the more developed countries), non-ferrous metal production and fossil fuel combustion.

Although lead does have minor natural sources (namely soil dust, volcanoes, forest fires, biogenic emissions and sea-salt spray) it has been shown that between 3700 and 5900 years ago, the atmospheric deposition rate was orders of magnitude lower than in the recent decades (Klaminder *et al*, 2003). It is therefore obvious that atmospheric Pb is today

primarily anthropogenic in origin. Furthermore, it has been suggested that, even in ancient times, atmospheric LRT dominated deposition in a Swedish bog (Klaminder *et al*, 2003). The analysis of Pb in a Greenland ice core provided evidence for hemispheric pollution by lead (Hong *et al*, 1994). Pollution of the Arctic by Pb over the last few decades has been ascribed, by isotope ratio analysis, largely to atmospheric LRT from Eurasian sources, especially Eastern Europe (Sturges and Barrie, 1989; Barrie *et al*, 1992). On the other hand, falling concentrations in ice cores in central Greenland in the latter part of the 20th century have been linked to declining emissions from North American sources (Rosman *et al*, 1993; McConnell *et al*, 2002).

Atmospheric inputs attributed to anthropogenic emissions from southern hemisphere sources have been identified in 20th century Antarctic snow and ice profiles (Rosman *et al*, 1994; Vallelonga *et al*, 2002; Planchon *et al*, 2003; van de Velde *et al*, 2005).

It has been suggested that lead deposition in western Ireland may have been largely due to atmospheric LRT from North America (Schell *et al*, 1997).

Steinnes *et al* (2005a) analysed the origin of Pb deposited in Norway from 1977 to 2000, showing that different parts of the country were affected by different source regions (western/central Europe, the U.K., Eastern Europe and Russia) and that the relative importance of these sources varied over the time period studied.

Interestingly, lead is one of the most ancient anthropogenic pollutants contributing to remote pollution through atmospheric LRT. Analysis of “varved” (annually layered) sediments in Sweden shows pollution beginning approximately 3000-4000 years ago (corresponding to the birth of metallurgy), with a peak in Roman times (about 2000 years ago) and a large and permanent Medieval increase (1000 years ago), arising from the development of metal production. Subsequently, there was a rapid increase after 1945, with a peak in the 1970s followed by the large decline, due to the use and subsequent phase-out of leaded petrol, (Brännvall *et al*, 1999, 2001; Renberg *et al*, 2000, 2002). Similar conclusions have been drawn from the study of cores taken from peat bogs in Spain (Martínez-Cortizas *et al*, 1997), Switzerland (Weiss *et al*, 1997; Shotyk *et al*, 1998) and Norway (Steinnes, 1997, 2005a, 2005b), although the Pb profiles may be influenced by local emissions as well as by atmospheric LRT. The history of atmospheric Pb deposition in a Swiss peat bog, going back over 12,000 years, has been deciphered by Shotyk *et al* (1998) and commented on by Nriagu (1998). A similar analysis of sources, covering the period from 3,000 to 500 years ago, has been provided by Hong *et al* (1994) from observations on Pb in a Greenland ice core. Determinations performed on Antarctic ice cores show “natural background” Pb concentrations until the late 19th century (Vallelonga *et al*, 2002). This demonstrates that the forms of Pb emitted from medieval and earlier metalworking sources in Europe and the Middle East were not long-lived enough in the atmosphere to cross the intertropical convergence zone and reach high southern latitudes.

2.3.3 Volatile organic halogen compounds

A number of volatile organohalogens have been mentioned above as being of partly natural and partly anthropogenic origin. Others are primarily or solely man-made. These include:

- chlorofluorocarbons (CFCs);
- CFC replacement compounds: hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs);
- halons and their substitutes;
- perfluorocarbons (PFCs);
- carbon tetrachloride (CCl₄);
- certain chlorinated solvents: methylene chloride (CH₂Cl₂), trichloroethylene (C₂HCl₃) and perchloroethylene (C₂Cl₄) (all of which may possibly have significant natural sources, although the evidence is not unequivocal: see Khalil *et al*, 1999; Moore, 2003, 2004), as well as the purely anthropogenic 1,1,1-trichloroethane (CCl₃CH₃).

The atmospheric lifetimes of these compounds range from about 5 days for C₂HCl₃ to thousands of years for perfluorocarbons (UNEP, 2002a), so all are liable to undergo LRT, as is demonstrated by regular monitoring performed at observation stations worldwide (WMO, 2002; Thompson *et al*, 2004). Nevertheless, only those substances that have atmospheric lifetimes greater than about one year will become more or less uniformly

distributed throughout the global atmosphere when they are emitted essentially only in the northern hemisphere. The shorter-lived compounds are not able to cross the intertropical convergence zone that acts as an atmospheric “barrier” close to the equator (Ballschmiter, 1992).

All these compounds are too volatile and insoluble in water for them to be substantially deposited from the atmosphere to ground level. Accordingly, they will in general have no direct impact on the biosphere following atmospheric LRT, their concentrations in background air being too low to affect vegetation, etc.. However, the parent compounds may well contribute to atmospheric impacts, such as stratospheric ozone depletion or global warming. Furthermore, their degradation products may be deposited to the biosphere. This is the case for the hydrohalic acids arising from most of the compounds listed above, while haloacetic acids can be formed in certain cases (e.g. trifluoroacetic acid from some HCFCs and HFCs, and trichloroacetic acid as a minor product from C₂Cl₄).

2.3.4 *Persistent organic pollutants (POPs) / Persistent, bioaccumulative and toxic substances (PBTs) / Semi-volatile organic compounds (SVOCs)*

A group of substances that has gained considerable attention in recent years is that of the “persistent organic pollutants” (POPs) or “persistent, bioaccumulative and toxic substances” (PBTs/PTBs). Definitions vary somewhat from one authority to another, but these terms are more or less synonymous and the acronym “POPs/PBTs” will be used henceforth in this dossier. Generally speaking, the definition is considered to encompass only substances that are deposited to the biosphere, where they may cause toxic effects, so volatile compounds such as the CFCs, while being persistent and organic, do not fall within this category.

The compounds that have been targeted up to present for action as priority substances in the major international regulatory agreements (UNECE and UNEP Conventions), as will be discussed later in this dossier, are:

- Polychlorinated dibenzodioxins (PCDDs) and dibenzofurans (PCDFs), and polycyclic aromatic hydrocarbons (PAHs). These substances are not produced intentionally, but trace amounts are formed during natural processes and anthropogenic activities (combustion, metallurgical operations, etc.);
- Polychlorinated biphenyls (PCBs) are no longer produced in the developed countries, but are still present in existing equipment as transformer fluids, etc.;
- Certain chlorinated pesticides: aldrin, chlordane, chlordecone, dieldrin, DDT, endrin, hexachlorocyclo-hexane isomers (HCHs) including lindane (γ -HCH), heptachlor, mirex, toxaphene (a mixture of chlorinated camphenes) and pentachlorophenol. Most of these have been phased out in the developed world but still persist in the environment;
- Hexachlorobenzene, a by-product formed in the manufacture of certain organic chlorine compounds, as well as being used formerly as a pesticide;
- Hexabromobiphenyl, a fire retardant.

The UNECE and UNEP lists are not definitive: new compounds are likely to be added in the future.

Since the substances mentioned above, in addition to being persistent, are in general hydrophobic and lipophilic, they tend to bioaccumulate in fatty tissues of living organisms from different levels in the food chain.

Many POPs/PBTs have such a low vapour pressure that a significant fraction of the total atmospheric concentration is present as material adsorbed on (or absorbed in) aerosol particles. Such species are often known as “semi-volatile organic compounds” (SVOCs). The partitioning between phases impacts the LRT potential, as will be discussed later in this dossier.

Transport to the polar regions

The persistence of the POPs/PBTs allows them to travel to remote areas of the planet, far from known sources. Many studies have focused on contamination of the Arctic, which is devoid of significant pollution sources of its own so that POPs/PBTs found there provide a convincing demonstration of transport from afar (Gregor and Gummer, 1989; Barrie *et al*,

1992; Macdonald *et al*, 2000a; AMAP, 1998, 2002, 2004; CACAR, 2003a; Bottenheim *et al*, 2004).

Depending on the location of the receptor site and the season, semi-volatile organochlorines and PAHs reaching the Arctic may have originated in Europe, Russia, East Asia or North America (Pacyna and Oehme, 1988; Halsall *et al*, 1997, 1998, 2000; Stern *et al*, 1997; Bailey *et al*, 2000; Macdonald *et al*, 2000a; CACAR, 2003a; Helm *et al*, 2004). Evidence has been presented that an episode of "brown snow" in the Canadian Arctic was due to emissions in China. The organic pollutants deposited included PAHs, PCBs, DDT-related compounds, toxaphene and the insecticides methoxychlor, endosulfan and HCH (Welch *et al*, 1991).

A number of modern agrochemicals, including trifluralin (short-lived in the atmosphere), have been found within the Arctic Circle (Welch *et al*, 1991; Chernyak *et al*, 1996; CACAR, 2003a), while chloroalkyl phosphate flame retardants and chlorobenzenes were identified in ice from a glacier in northern Sweden (Laniewski *et al*, 1998).

Pollution of the Antarctic by a range of organochlorines has also been demonstrated (Larsson *et al*, 1992; Kallenborn *et al*, 1998; Laniewski *et al*, 1998; Corsolini *et al*, 2002; Borghini *et al*, 2005; Dickhut *et al*, 2005).

Transport to other regions

The herbicide atrazine is known to be transported through the atmosphere over distances of hundreds of kilometres (Thurman and Cromwell, 2000).

On a more regional scale, atrazine and related compounds detected in the pristine Isle Royale National Park in Lake Superior were assigned sources in Minnesota, Iowa and Wisconsin (Thurman and Cromwell, 2000), while organophosphate pesticides applied in California's Central Valley were shown to be transported to the Sierra Nevada mountains (Zabik and Seiber, 1993; Aston and Seiber, 1997).

Transpacific transport from Asia to the North American west coast of a number of pesticides (including HCHs, endosulfans, dacthal, chlorothalonil and trifluralin), as well as the more volatile PCBs, has been described in several studies (Koziol and Pudykiewicz, 2001; Wilkening, 2001; Killin *et al*, 2004; Lichota *et al*, 2004; Harner *et al*, 2005).

A number of pesticides that are not authorised for use in Denmark have nevertheless been reported by Asman *et al* (2005) to be present in precipitation there. They are therefore assumed to have been transported via air from other countries.

Toxaphene, formerly used as a cotton insecticide in the southern United States, has been shown to be transported through the atmosphere to Bermuda, western Canada and the Great Lakes (Bidleman and Olney, 1975; Donald *et al*, 1998; James and Hites, 2002).

PCBs in the air above the eastern Mediterranean, as well as PAHs in marine sediments there, have been demonstrated to originate in western and central Europe (Mandalakis and Stephanou, 2002; Tsapakis *et al*, 2003).

The modelled contribution of transboundary transport to air concentrations and deposition fluxes, in various European countries and the Arctic, of benzo[a]pyrene, PCDD/Fs, PCBs and γ -HCH is discussed by Shatalov *et al* (2004).

The observation of the latitudinal dependence of the concentrations in air of a number of organochlorine pesticides and related compounds has been used to determine empirical "travel distances" for these substances from their points of release at various locations in North America. The values range from hundreds to thousands of kilometres (Shen *et al*, 2004, 2005). Travel distances of comparable magnitudes were determined for currently used pesticides belonging to various chemical families, by analysing the latitudinal dependence of their concentrations in North American lakes (Muir *et al*, 2004).

3 Long-Range Transport through media other than the atmosphere

Although the atmospheric pathway is believed to be the dominant one for the LRT of chemicals and is the one which has been the subject of the most scientific studies, it is not the only one. In this chapter, three other modes of LRT are discussed.

3.1 Transport by Rivers, Oceans and Drifting Ice

Some substances may not be prone to atmospheric LRT, but may nevertheless be transported over long distances, by rivers and the oceans. This is the case, in particular, for compounds that, on account of their high affinity for aqueous phases:

- are emitted to water and remain there;
- are emitted initially to air, but subsequently partition to a greater or lesser degree to water.

The first of these cases can be illustrated by transport to the Arctic Ocean of artificial radionuclides, especially ^{137}Cs and ^{129}I , discharged into the Irish Sea and the English Channel from nuclear fuel reprocessing facilities at Sellafield (U.K.) and La Hague (France), respectively. The emissions from these two point sources have been well characterised and observations of the resulting radioactivity have provided scientists with valuable information on ocean circulation. The time-scale for oceanic transport from the U.K. and French plants to the Norwegian Sea and to the Arctic Ocean and its marginal seas is of the order of 5 years (Smith *et al*, 1990, 1998; Kershaw and Baxter, 1995; CACAR, 2003a; Gao *et al*, 2005), compared to only a few days for atmospheric transport from mid-latitudes to the Arctic.

For many water-soluble species, including nitrates and dissolved organic matter, the flow from the continents to the oceans is primarily due to their transport in groundwater, streams and rivers (Wörman, 1998; Kaiser, 2002; Bouwman *et al*, 2005). Thus, rivers export to the oceans about 50 million tons per year of dissolved nitrogen, arising from natural sources as well as from agriculture, sewage and industry (Bouwman *et al*, 2005). Rivers are also a prominent source of particulates and sediment to the world's oceans (Depetris, 2000; Syvitski *et al*, 2005). Nuclear weapon plants located along Siberian rivers have been reported to contribute to pollution of the Arctic by radionuclides (Livingston, 1995).

The case of substances initially emitted to air, but subsequently undergoing significant oceanic transport, could no doubt be exemplified by highly water-soluble compounds such as the inorganic acids HCl and HF. It is indeed reasonable to assume that these species, after being removed from the atmosphere by wet deposition, will end up in rivers and the oceans, where their concentrations will become evened out by mixing. However, this phenomenon would be difficult to demonstrate, on account of the permanent background concentrations of the corresponding anions in seawater. It is nevertheless known that riverine transport plays a major role in the transport of anthropogenically "fixed" nitrogen species (mainly ammonium and nitrate ions) to marine ecosystems (Galloway *et al*, 1995).

Macdonald *et al* (2000b) predict that the herbicide atrazine, on account of its high solubility and long lifetime in water, will behave much like ^{137}Cs in the ocean, i.e. providing a tracer of water from drainage basins where it has been applied.

Although polychlorinated hydrocarbons are hydrophobic, some of them have solubilities in water great enough for the oceans to play a role in their LRT, even if the atmosphere remains the primary vector. That is to say, dissolution in seawater, particularly in the colder regions of the planet, can lead to a significant marine burden. The dissolved species can then become geographically redistributed in the oceans, while undergoing a continuous exchange with the atmosphere. This phenomenon has been discussed, for example, for the HCHs, DDTs, PCBs, chlordanes, etc. (Iwata *et al*, 1993, 1994; Schreitmüller and

Ballschmiter, 1994; Macdonald *et al*, 2000a, 2000b; Koziol and Pudykiewicz, 2001; Scheringer and Wania, 2003; CACAR, 2003a).

A large reservoir of α -HCH has accumulated in the Arctic Ocean, mainly on account of atmospheric transport from lower-latitude regions where the “technical” HCH mixture (of which α -HCH is the major component) was used as a pesticide in the 1970s and 1980s. The accumulated aqueous reservoir will ultimately be shed predominantly through water currents and degradation, since extensive ice cover prevents effective exchange with the atmosphere, despite the fact that the ocean water is now oversaturated with α -HCH relative to air (Wania and Mackay, 1999; Wania *et al*, 1999; Macdonald *et al*, 2000a, 2000b; Scheringer and Wania, 2003; CACAR, 2003a).

Input of PCBs, HCHs and DDTs to the Arctic Ocean via LRT in Russian rivers may have been significant in the past and may still be so today (Wania and Mackay, 1999; Macdonald *et al*, 2000a, 2000b; AMAP, 1998, 2002, 2004; CACAR, 2003a). Several large rivers flowing through industrialised and agricultural areas drain into the Arctic Ocean. When their sediments reach the estuary, they are partially incorporated into sea ice. Ice-borne particles, possibly laden with contaminants, may thus be released when the ice melts after drifting to other parts of the ocean (AMAP, 1998, 2002, 2004).

However, Gustaffson *et al* (2005) present evidence showing that drifting ice is not an important LRT carrier of POPs to the Arctic marginal ice zone.

3.2 Transport by migrating animals

As pointed out by Wania (1998), certain migrating species cover long distances, often crossing international boundaries and linking industrialised or agricultural regions with remote ones. Furthermore, persistent lipophilic substances can bioaccumulate in animal tissues – particularly in those at the top of the food chain – to levels that are orders of magnitude greater than in air or water.

Unlike atmospheric LRT, which ultimately leads to deposition over broad expanses of the Earth’s surface, transport of contaminants concentrated in animal tissues can potentially provide a focused delivery to a given ecosystem.

A preliminary analysis of the significance of LRT by migratory seabirds and whales was performed by Wania (1998) for selected POPs/PBTs (HCHs, PCBs and DDTs). From this assessment, one may conclude that:

- Gross transport rates of POPs/PBTs via these animals into a given region are usually smaller than the fluxes in air or water, but in some cases may be of a comparable order of magnitude;
- It is more difficult to assess whether or not net transport (e.g. import into the Arctic minus export from the Arctic) actually occurs. This depends on the extent to which the animals feed, excrete, spawn or die in distinct areas.

One case has received particular attention, namely that of salmon making their end-of-life migration from the northern Pacific Ocean to their natal freshwater lakes in Alaska, to spawn and die. The decomposing carcasses of the salmon release PCBs, DDTs and other POPs/PBTs accumulated over their lifetime, so that the sediments of these lakes and the other fish that inhabit them can become contaminated with particularly elevated levels of these contaminants (Ewald *et al*, 1998; Krümmel *et al*, 2003, 2005; Mu *et al*, 2004). Indeed, the PCB input flux to the lakes by this “biological pump” mechanism can be up to sixfold greater than atmospheric deposition (Krümmel *et al*, 2003).

Salmon have also been shown to be a significant local vector for mercury, in the readily bioavailable form of monomethylmercury, to Alaska’s water bodies (Zhang *et al*, 2001).

It has been hypothesised that deposited seabird guano may influence the levels of PCBs and other POPs/PTBs on isolated islands in the Norwegian Arctic (Enge *et al*, 2001; AMAP, 2002, 2004; Evenset *et al*, 2004, 2005) and in the Canadian Arctic (Blais *et al*, 2005). This may also be true for certain radionuclides, but the evidence is equivocal (Dowdall *et al*, 2005).

3.3 Anthropogenic transport

When a xenobiotic substance is found in a reputedly pristine environment, one should always bear in mind the possibility that it may have reached that location, partially or wholly, through “anthropogenic” transport, that is the shipment of goods or hazardous wastes (including ocean dumping of the latter), or the displacement of persons, for trade, tourism, or other purposes. Even the polar regions have a small resident or temporary population (indigenous peoples, military personnel, oil industry workers, scientific researchers, explorers, etc.) who may possibly use articles or products containing the substances concerned. Local anthropogenic contamination of the Arctic (by airstrips, military installations, mining, industry, waste dumps, etc.) has been reviewed by CACAR (2003a). Stow *et al* (2005) have quantified the contribution of Distant Early Warning radar stations to local and Arctic-wide contamination by PCBs.

There are regrettably numerous examples of accidental leakage of anthropogenically transported substances that have led to significant environmental contamination and damage, e.g. from oil tankers, nuclear-powered submarines, etc. Illegal dumping of waste materials into the oceans is also still a major concern.

4 Mechanisms and modelling of atmospheric Long-Range Transport

4.1 Pathways and timescales

The atmosphere clearly provides a major vector for the global dispersion of pollutants. The pathways followed and the distances covered depend on the physical properties and chemical reactivity of the substances considered, as well as on the location and moment (season and time of day) of release to the atmosphere and on the prevailing meteorological conditions. Compounds that degrade rapidly in the atmosphere will, in general, not travel as far as more persistent ones. Substances with very low volatility will tend to adsorb onto atmospheric particles, which, depending on their size, may settle rapidly to the ground. Hydrophilic pollutants will be removed by precipitation more readily than less soluble ones.

The observational evidence presented in Chapter 2 of this dossier clearly shows that there are preferential (albeit to some extent seasonally-dependent) pathways for intercontinental atmospheric LRT, e.g. eastwards from Asia to North America and thence to Europe, northwards from Eurasia to the Arctic, westwards from northern Africa across the Atlantic, south-eastwards from southern Africa to Australia and New Zealand, etc..

The meteorological phenomena driving these pathways have recently been reviewed by Stohl *et al* (2002a), EMEP (2002), Holloway *et al* (2003) and Stohl (2004). Various individual chapters in the book by Stohl (2004) deal with atmospheric pollutant transport between most of the major regions of the world, namely the Arctic (Bottenheim *et al*, 2004), North America (Cooper and Parrish, 2004), Europe (Huntrieser and Schlager, 2004), East Asia (Pochanart *et al*, 2004), Southern Asia (Lawrence, 2004) and Africa (Piketh and Walton, 2004).

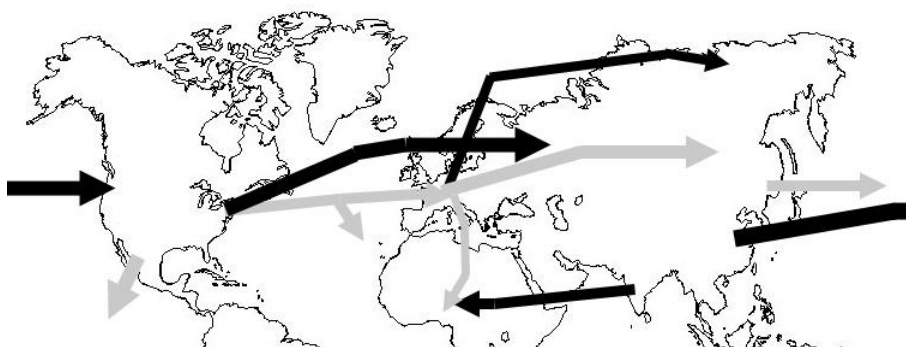
The preferential pathways for atmospheric LRT in the northern hemisphere are illustrated schematically in Figure 1 (adapted from Stohl, 2004).

Further reviews of the pathways by which pollutants are transported through the atmosphere specifically into the Arctic region are provided by Barrie *et al* (1992), Mackay and Wania (1995a), Iversen (1996), AMAP (1998, 2004), CACAR (2003a) and Macdonald *et al* (2000a, 2005). The effect of the North Atlantic Oscillation (NAO) on air pollutant transport into the Arctic has been discussed by Eckhardt *et al* (2003), MacLeod *et al* (2005) and Hermanson *et al* (2005). The NAO, the intensity of which is determined by the difference between the subtropical high pressure and the polar low pressure regions, is the dominant driver of climate variability over the North Atlantic.

For the express purpose of assessing the ability of various models to represent atmospheric dispersion and transport pathways, inert synthetic tracer gases have been released to the atmosphere in small quantities. For instance, perfluoromethylcyclohexane was released in 1994 from a location in northwestern France and tracked at 168 sampling locations in 17 countries across Europe (Nodop *et al*, 1998; Stohl *et al*, 1998; Warner *et al*, 2004, 2005). Similar experiments had previously been conducted in North America (Draxler, 1991; Sykes *et al*, 1993; Stohl *et al*, 1998; Draxler and Hess, 1998). The monitoring of “tracers of opportunity” resulting from deliberate or accidental releases of substances such as radionuclides, has also been used for model validation.

Intercontinental transport occurs on time-scales of about 3-30 days (Sturman *et al*, 1997; Stohl *et al*, 2002a) and is thus relevant for compounds having a lifetime of at least 3 days (corresponding to a half-life of 2 days: see Chapter 7 below). This includes substances such as ozone and many of its precursors, certain aerosols, mercury and POPs/PBTs (Stohl *et al*, 2002a), as well as a range of volatile organic halogen compounds, as discussed above. Under exceptional meteorological circumstances, “intercontinental pollution express highways” may arise, leading (for example) to transport across the Atlantic in as little as one day (Stohl *et al*, 2003a).

Summer



Winter

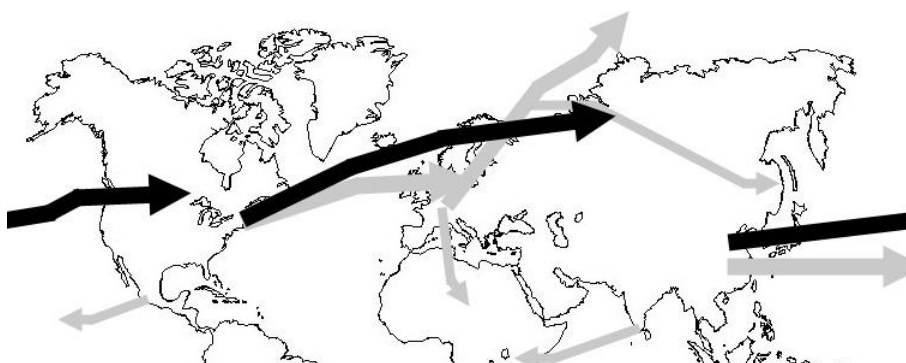


Figure 1: Grey arrows depict transport in the lower troposphere (below 3km), while black arrows refer to the middle and upper troposphere (above 3km).

Air quality at a given location can be influenced by emissions from another continent either through an increase in the more or less ubiquitous hemispheric background levels of pollution, or through discrete episodic flows of enhanced pollutant levels. The contributions from the latter events depend on location, season and the substance considered.

Intercontinental transport episodes may involve either (a) advection more or less horizontally within the planetary boundary layer; or (b) lifting into the free troposphere via deep convection, “orographic effects” (i.e. the influence of mountain slopes on air flow) or, in particular, large-scale weather systems, especially the uplifting “warm conveyor belts” that occur ahead of advancing cold fronts. The relative importance of these phenomena differ from region to region and from substance to substance (EMEP, 2002; Stohl *et al*, 2002a; Holloway *et al*, 2003; Stohl, 2004).

The distance over which LRT occurs thus depends greatly on meteorological conditions (wind speed, precipitation and the orographic and frontal effects mentioned above) as well as on the properties of the substance itself (reactivity, solubility and volatility). Pollutants transported in the free troposphere will typically be carried further and faster than those in the boundary layer, owing to stronger winds and fewer loss mechanisms (Holloway *et al*, 2003).

From Asia to North America and from North America to Europe, transport in the free troposphere appears to be more important than advection in the boundary layer. Between Europe and Asia, boundary-layer transport and orographic lifting may be dominant (EMEP, 2002).

4.2 “Global fractionation” and the “grasshopper effect”

Rappe (1974) suggested that pesticides “when used in warm climates, become evaporated and transported to cool areas where they will be condensed”. Goldberg (1975) coined the term “global distillation” to describe the process whereby a compound could volatilise from warmer regions, undergo long-range atmospheric transport and subsequently “recondense” in colder areas at higher latitudes. Ottar (1981) postulated “a systematic transfer of the

more persistent [organochlorine] compounds from warmer to colder regions”, leading eventually “to an accumulation of these substances in the temperate and Arctic regions”.

Wania and Mackay (1993a) suggested that, through “global distillation”, organic compounds could become latitudinally fractionated, “condensing” at different temperatures according to their volatility, so that compounds with vapour pressures in a certain low range might accumulate preferentially in polar regions.

It should be emphasised that even organic pollutants with very low volatilities do not actually condense – even at the lowest environmental temperatures – in the sense that the atmosphere becomes saturated or supersaturated. Indeed, atmospheric partial pressures are always below the vapour pressures. Rather, cold conditions favour partitioning from air to a non-gaseous phase, which is the meaning of the term “cold condensation” as used by Wania and Mackay (1993a).

Wania and Mackay (1996) and Wania (1999) analysed the factors involved in global fractionation. Firstly, cool temperatures favour greater adsorption to atmospheric particulate matter, which is then deposited to the Earth’s surface. Secondly, natural decomposition also slows down in the cold, making pollutants more persistent. Thirdly, low temperatures reduce the rates of evaporation of contaminants from water and promote their partitioning from the atmosphere to the surface soil or vegetation.

According to Wania and Mackay (1996), the global distribution of organic pollutants will depend largely on their vapour pressure (or their octanol-air partition coefficient) and the ambient temperature.

The migration of semi-volatile compounds to higher latitudes, in a series of cycles of atmospheric transport, deposition and re-volatilisation, in tune with seasonal temperature changes, has come to be known as the “grasshopper effect” (Wania and Mackay, 1996).

The concept of “global fractionation” and observational evidence for it were critically analysed by Ockenden and Jones (1999). More recent evidence for the “grasshopper effect” has been reviewed by Gouin *et al* (2004). Some of the environmental models to be discussed below enable one to calculate the number of “hops” between mobile and non-mobile media that are associated with a given journey from warm to cold regions (e.g. Semeena and Lammel, 2003, 2005; Leip and Lammel, 2004).

A number of different environmental matrices have been analysed as a function of latitude, in order to provide evidence for the global fractionation theory, including air (Iwata *et al*, 1993; Ockenden *et al*, 1998a; Agrell *et al*, 1999; Meijer *et al*, 2003a), river water (Iwata *et al*, 1994), seawater (Iwata *et al*, 1993; Wania and Mackay, 1996), soil (Lead *et al*, 1997; Meijer *et al*, 2002, 2003b; Ockenden *et al*, 2003), sediments (Iwata *et al*, 1994; Muir *et al*, 1996), tree bark (Simonich and Hites, 1995), various plants (Calamari *et al*, 1991; Ockenden *et al*, 1998b) and frogs (ter Schure *et al*, 2002).

In a similar manner to the temperature-driven latitudinal distribution of semi-volatile compounds, concentration gradients have been observed in mountainous areas as a function of altitude (for reviews, see Fernández and Grimalt, 2003; Daly and Wania, 2005).

In connection with the temperature-dependent partitioning between mobile and non-mobile media, Ockenden *et al* (2003) have shown that the forested soils of the northern hemisphere, and other carbon-rich soils, appear to be playing an important role in “protecting” the Arctic from LRT of PCBs (for example) through their absorptive capacity for these compounds. Thus, in the 70 years since PCBs were first produced, only a very small fraction of the total inventory has travelled from the populated source regions to the Arctic, mainly on account of retention by soils (Ockenden *et al*, 2003). Vegetation may have a similar effect, but its role is complex (Dalla Valle *et al*, 2004).

4.3 Atmospheric transport of substances of marine origin via sea-salt aerosols

Certain natural substances and anthropogenic pollutants present in the oceans, especially those with surfactant properties, become concentrated in the sea-surface microlayer,

having been brought to the surface by the scavenging action of ascending air micro-bubbles entrained underwater by breaking waves or falling rain. The marine aerosols that are formed when the air bubbles burst may be highly enriched in surfactants and other substances (e.g. certain metal ions) that interact with them, as well as microorganisms (Marty *et al*, 1979; Hardy, 1982; Blanchard, 1989; Tseng *et al*, 1992; Skop *et al*, 1994; GESAMP, 1995; Stefan and Szeri, 1999; Oppo *et al*, 1999; Grammatika and Zimmerman, 2001; Mochida *et al*, 2002; Saint-Louis and Pelletier, 2004). This phenomenon provides a mechanism for efficient sea-to-air transfer even for certain substances with low volatility and high water solubility, potentially enabling their LRT via the atmosphere (Cini and Loglio, 1997).

4.4 Relationship between long-range transport and persistence

LRT is clearly related to persistence in the environment, since time is required for transport. A long-lived chemical will travel further than a more reactive one with similar physico-chemical properties other than persistence. However, the propensity for LRT cannot be predicted from persistence alone. Even highly persistent chemicals will not undergo atmospheric LRT if their volatility is very low and/or their aqueous solubility is high. Furthermore, the societal concerns related to persistence and LRT are fundamentally different: persistent chemicals may adversely affect future generations, while those undergoing LRT are liable to contaminate areas where they have not been used (van de Meent *et al*, 2000).

4.5 Models

A huge number of numerical models, covering a wide range of complexity, have been developed for predicting how chemicals are transported, distributed and degraded in the various compartments of the environment. It is beyond the scope of this dossier to attempt to discuss or even list them all. Some useful overviews have been provided by AMAP (1998, Chapter 3), Mackay *et al* (2001), Gouin *et al* (2001), Scheringer and Wania (2003), Schultz and Bey (2004) and OECD (2004).

It is important to recognise that POP/PBT-type compounds – on which this chapter on modelling is focused – are “multimedia” chemicals. That is to say they are liable to be present simultaneously in the various environmental media or compartments, such as air and the particles it contains, surface and ocean water, soil, sediments and vegetation, and they cycle between these different media. Any model used for describing the environmental behaviour of POPs/PBTs, including their potential for LRT, must therefore take into account intermedia exchange as well as transport and degradation in the various compartments.

Regional, hemispheric or global 3-dimensional chemistry-transport “general circulation” models (GCMs), with high spatial resolution and carefully validated by observations, are perhaps the only adequate tools for satisfactorily describing how emissions at one location may affect another location thousands of kilometres away. Such “dispersion” models, initially developed for other classes of substances, have been applied to POP/PBT-type compounds by Jacobs and van Pul (1996), van Jaarsveld *et al* (1997), Lammel *et al* (2001a, 2001b), Semeena and Lammel (2003), Leip and Lammel (2004), Koziol and Pudykiewicz (2001), Malanichev *et al* (2002, 2004), Ma *et al* (2003, 2004), Hansen *et al* (2004) and Suzuki *et al* (2004).

However, less computationally-demanding multimedia compartment-based models (or “box” models), with a lower spatial resolution, are very useful tools for screening and ranking purposes. They are based on the fugacity modelling concept originally developed by Mackay and coworkers in their suite of zero-dimensional CEMC Level I, II, III and IV models (for an overview, see Mackay, 2001). This concept has been adapted and in some cases extended, leading to a series of models, many of which are spatially resolved: EQC (Mackay *et al*, 1996), the Bergen model (Strand and Hov, 1996), GloboPOP (Wania and Mackay, 1993a, 1993b, 1995, 1996, 1999, 2000a; Wania, 2003), CoZMo-POP (Wania *et al*, 2000), CalTOX (McKone and Enoch, 2002), SimpleBox (van de Meent, 1993; Brandes *et al*, 1996), EUSES (derived from SimpleBox and adopted for European Union risk assessment purposes, see Vermeire *et al*, 1997 and <http://ecb.jrc.it/>), ChemRange (Scheringer, 1996, 1997, 2002; Scheringer *et al*, 2004a, 2004b), CliMoChem (Scheringer *et al*, 2000, 2004a), ELPOS (<http://www.usf.uos.de/projects/elpos/>; Beyer and Matthies,

2001), TaPL3 (<http://www.trentu.ca/cemc/models/TP300.html>; Beyer *et al*, 2000), BETR (MacLeod *et al*, 2001; Woodfine *et al*, 2001; Prevedouros *et al*, 2004; Toose *et al*, 2004), IMPACT 2002 (Pennington *et al*, 2005); and others.

The following sections focus on (a) the modelling studies performed in order to define the LRT potential of chemicals (especially POPs/PBTs) released to the atmosphere; and (b) source-receptor relationships.

4.6 Long-range transport potential

Many of the international regulatory agreements on the abatement of transboundary pollution explicitly include the potential for LRT as one of the criteria to be taken into consideration for control of a given chemical (see Chapters 6 and 7 below). However, none of the agreements currently provide a definition according to which the LRT potential itself can be expressed in numerical terms. Nevertheless, in support of possible future regulatory action, a number of groups of scientists have applied mathematical models to derive quantitative measures of the LRT potential (LRTP).

The development of the LRTP concept has been carried out mainly using multimedia compartment-based models in the hypothetical “evaluative” mode. The intention in this case is not to describe three-dimensional transport referenced to actual geographical coordinates and adopting realistic scenarios for emissions from defined locations. The models are merely used to explore in a generic way how the LRTP depends on substance properties, environmental properties, mode of entry into the environment, etc., and to assist substance evaluation, for instance by providing rankings of different compounds.

The LRTP has been expressed by different authors in terms of “spatial range (SR)” (Scheringer and coworkers), “characteristic travel distance (CTD)” (McKone, Matthies and coworkers), or similar descriptors. For overviews of these somewhat differing concepts, see Scheringer *et al*, 2001; Mackay *et al*, 2001; Scheringer and Wania, 2003.

The early work on LRTPs was reviewed by van de Meent *et al* (2000), who discussed three alternative derivations, based on the following approaches:

1. The “mixed compartment” approach: A multimedia box model is used to simulate a steady-state open system, in which the compartments are all well-mixed and the size of the system is just great enough for overall transformation flux of the compound in the various media to be equal to its advection flux out of the atmospheric compartment, each of these terms representing 50 % of the input into the system (see Figure 2). According to this approach, the characteristic scale or characteristic travel distance (CTD) is given by $L = u_A \cdot \tau \cdot F_A$ (van de Meent *et al*, 2000; Beyer *et al*, 2000), where u_A is the air compartment advection velocity (i.e. wind speed), τ is the overall multimedia lifetime or persistence (as defined by Bennett *et al*, 1999) and F_A is the fraction of the chemical present in the air compartment. The EQC model may be used for such calculations (Beyer *et al*, 2000);

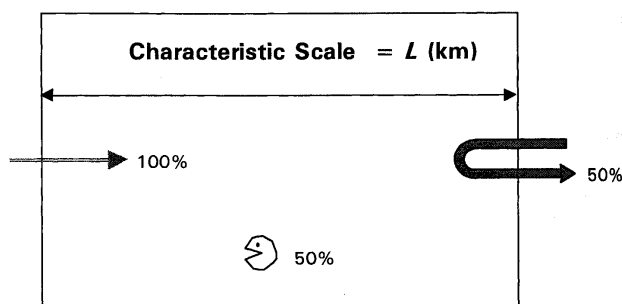


Figure 2: Schematic representation of an open multimedia box model proposed for calculating the “characteristic scale” as a measure of LRT potential (from van de Meent *et al*, 2000)

2. The “Lagrangian” approach: The travel distance of a parcel of a mobile compartment (air or water) is calculated, such that a specified percentage of the initial amount of a compound in that moving cell is lost as the result of transformation and intermedia transport. This calculation is performed using a “Lagrangian” model (i.e. one in which composition changes are described relative to the moving fluid parcel, as opposed to the “Eulerian” approach of the box models, in which the coordinates are fixed in space). This Lagrangian approach considers the movement of a compound driven by advective flow (e.g. in air) over a non-mobile phase such as soil or vegetation. During transport, the concentration in the moving phase decreases as a consequence of transformation in each compartment and intermedia mass exchange. At steady state, there is a unique distance from the emission source at which the concentration has fallen to 37 % (1/e) of its initial value. This is the characteristic travel distance (Bennett *et al*, 1998) or travelling distance (van Pul *et al*, 1998), as illustrated in Figure 3. The basic assumptions involved are listed by van de Meent *et al* (2000);

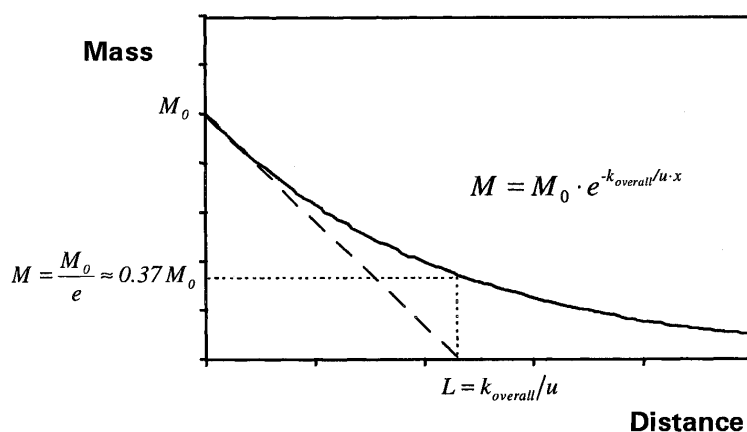


Figure 3: The concept of characteristic travel distance in the Lagrangian approach (from van de Meent *et al*, 2000)

3. The “Eulerian multi-box” approach: In the ChemRange model (Scheringer, 1996, 1997, 2002; Scheringer and Wania, 2003; Scheringer *et al*, 2004a), the Earth is divided into a one-dimensional loop of interconnected 3-compartment latitudinal zones that exchange air and ocean water via dispersive eddy diffusion (rather than advection as in the two previous approaches), as illustrated in Figure 4. A chemical is released to one of the zones and the substance undergoes transport (driven by the concentration gradient), degradation and partitioning. The spatial range (SR) of the compound is defined as the distance that incorporates 95 % of area under the curve representing concentration as a function of distance from the source. The calculation can be performed for pulse or steady-state releases of chemicals. A more realistic geometry, in which the Earth is represented by a variable number of interconnected, well-mixed latitudinal zones (CliMoChem model, see Figure 5), was later proposed by Scheringer *et al* (2000).

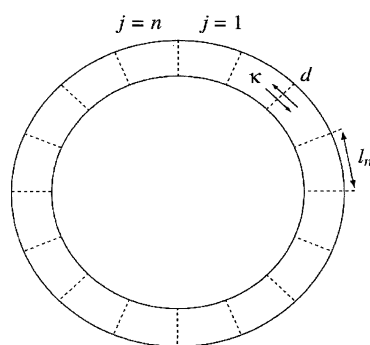


Figure 4: Geometry of the Eulerian multi-box ChemRange model used by Scheringer and coworkers (from van de Meent *et al*, 2000)

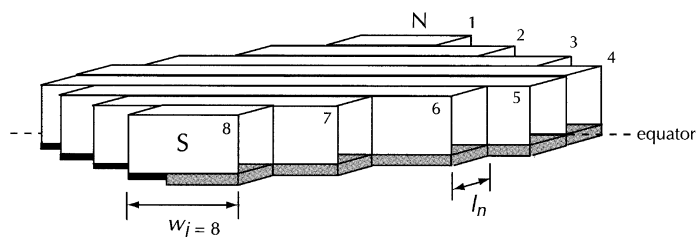


Figure 5: Geometry of the Eulerian multi-box CLiMoChem model used by Scheringer and coworkers (from Scheringer *et al*, 2000)

More recent work on the development of the concept of LRT potentials and the models used for deriving is presented in the following studies:

- The TaPL3 model has been developed specifically for calculating overall persistence and LRTP. It is based on the EQC model, but has no advective loss terms. It leads to an expression for the characteristic travel distance identical to that of approach 1 above (Beyer *et al*, 2000);
- Beyer *et al* (2000) introduced the concept of “stickiness”, i.e. the fraction of a substance that will partition into a non-mobile medium such as soil or vegetation and hence retard transport. Sweetman *et al* (2005) also investigated the impact of adsorption to soil on characteristic travel distance;
- Wania and Mackay (2000b) compared atmospheric travel distances calculated using various models. Although the absolute values obtained varied substantially from model to model, the relative rankings were similar. This led the authors to suggest the use of a benchmark substance, with the transport criterion being expressed in the following terms “A chemical shall be considered as having the potential for atmospheric long-range transport if its calculated transport distance/spatial range in a typical regional level III [i.e. open, steady-state] multimedia model exceeds that of chemical X calculated under the same conditions”. Wania and Mackay (2000b) also emphasised the fact that the LRTP depends on the mode of entry into the environment, i.e. the compartment into which the compound is emitted, as previously demonstrated for overall persistence (Webster *et al*, 1998). For instance, a substance deposited on to soil will be transported less readily through air than the same substance emitted directly to the atmosphere;
- Other comparisons of models, and of the concepts of CTD and SR, were provided by Bennett *et al* (2001), Beyer *et al* (2001), Wania and Dugani (2003) and Stroebe *et al* (2004);
- Beyer and Matthies (2001) investigated the effect of combined transport in air and water on the CTD;
- Pennington (2001) presented a tiered methodology for deriving atmospheric travel distances, for screening purposes, using a minimum of environmental degradation data;
- Wania (2003) extended the LRTP concept by introducing “immediate and long-term Arctic Contamination Potentials” (ACPs), defined as the fractions of the total amount of a chemical present in surface media that reside in the Arctic after 1 or 10 years of steady emissions with a generic global distribution. Wania (2003) showed how the calculated ACPs depend on substance properties by plotting them in a 2-dimensional “space”, the axes of which are the logarithms of the octanol-air and air-water partition coefficients (the latter being the adimensional Henry’s Law constant);
- Beyer *et al* (2003) investigated how temperature influenced CTD by modifying degradation rates and partitioning;
- Scheringer *et al* (2004b) showed that inclusion of export to the deep sea with settling particles could considerably reduce the SRs of highly hydrophobic chemicals, calculated using the ChemRange model (see also Dachs *et al*, 2002);
- Leip and Lammel (2004) investigated various LRTP indicators using a global, dynamic (non steady-state), geo-referenced, multi-compartment chemistry-transport model with high spatial and temporal resolution. The authors demonstrated that the absolute values of LRTP and even the substance-to-substance rankings varied greatly not only with the geographical direction considered (N-S or W-E), but also with the location of release on the Earth’s surface;

- Breivik *et al* (2004) used the CoZMo-POP model to show that seasonally varying environmental conditions (temperature, precipitation and atmospheric hydroxyl radical concentration) can cause the CTD to vary by a factor of ten or more over the course of a year. This corroborates the conclusion of Franklin *et al* (2000), namely that in winter months at mid to high latitudes, the hydroxyl concentration is so low that the vast majority of organic compounds have atmospheric half-lives greater than 2 days, a cut-off value proposed by several regulatory bodies as a criterion for determining propensity for LRT (see Chapter 7 below). Observational evidence provided by Hermanson *et al* (2005) also points to the effect of low reactivity in polar regions. A number of pesticides in current use having “global mean” atmospheric lifetimes of only hours are, nevertheless, found in an ice-core from the Norwegian Arctic. Their LRT from source regions is explained by low atmospheric OH concentrations and consequently enhanced persistence during winter months;
- Wania *et al* (2004) provided a useful analysis of the limitations and shortcomings of multimedia fate and transport models for predicting partitioning and LRTP of chemicals, particularly those that are very soluble in water and those that sorb strongly to atmospheric particles;
- Liu *et al* (2005) defined “transpacific transport potentials” as a function of the region of emission in Asia and atmospheric lifetime of the substance considered;
- As already mentioned above, empirical travel distances have been derived from observations on the latitudinal dependence of various families of chemicals in air (Shen *et al*, 2004, 2005) and water (Muir *et al*, 2004). The values obtained were compared to those calculated using various models;
- To address the need for critically examining the various existing models for determining persistence and LRTP, the OECD established an expert working group to assess their reliability and applicability (OECD, 2002, 2004). A recent paper (Fenner *et al*, 2005) by this group provides a comparison of nine models, using a set of 3175 hypothetical chemicals exhibiting a broad range of physico-chemical properties. It was concluded from this exercise that the rankings of persistence and LRTP are highly correlated among the models and are largely determined by the substance properties;
- Recently, Mackay (2005) has introduced the concept of “distant residence time (DRT)” as an indicator of LRTP. In a multi-box environmental model, the DRT is the proportionality constant at steady state between the inventory in a distant box and the discharge rate into the box where the compound is released.

4.7 Source-receptor relationships

Certain atmospheric models have been developed for the express purpose of exploring so-called “source-receptor” relationships. Such modelling is described as “receptor-oriented” if it is focused on the point of impact or, in other words, if it attempts to answer the question “At a given location, where do the pollutants come from?” On the other hand, modelling is regarded as “source-oriented” if it tracks contaminants away from a given source and aims at answering the question “Which downwind locations will be affected by these emissions?” Both questions are important yet sensitive ones, since they may have liability implications.

The receptor-oriented and source-oriented approaches involve establishing trajectories backwards to the source, or forwards from the emission point, respectively. Forward trajectories indicate where an air parcel will go, while backward trajectories describe where it came from. The impact at the receptor site may be expressed in terms of pollutant concentrations or deposition fluxes.

Trajectory models, which describe the paths taken by air parcels, are generally Lagrangian in nature since they are then computationally less demanding than purely Eulerian ones (Stohl, 1998; Seibert and Frank, 2004). However the hybrid Lagrangian-Eulerian model HYSPLIT (Draxler and Hess, 1998) has gained great popularity for studying source-receptor relationships, along with the Lagrangian models ATMOS (Arndt and Carmichael, 1995; Arndt *et al*, 1997, 1998), FLEXPART (Stohl *et al*; 1998, 2002b, 2003b; Stohl and Thompson, 1999) and STILT (Lin *et al*, 2003).

A detailed discussion of source-receptor relationships is beyond the scope of this dossier. A few illustrative, but far from comprehensive, examples of their use are nevertheless given below:

- Using HYSPLIT, Commoner *et al* (2000) estimated the amounts of PCDD/Fs – emitted by 44000 sources – that are deposited in several communities of the Canadian High Arctic. The authors concluded that a fairly limited number of North American sources are responsible for almost all of the PCDD/Fs deposited on the territory of Nunavut;
- Cohen (2001) similarly assessed the origins of PCDD/Fs deposited in the Great Lakes;
- Han *et al* (2005) used HYSPLIT to trace the origin of atmospheric reactive gaseous mercury at rural locations in New York State. Coal-fired power plants and mining and smelting operations in the eastern USA and Canada were implicated;
- Arndt *et al* (1998) used the ATMOS model to determine the source-receptor relationships for sulphur deposition in various Asian countries;
- Astitha *et al* (2005) described a methodology for identifying the origin of air pollutants observed at a remote location in the Eastern Mediterranean;
- FLEXPART has been used extensively by Stohl and coworkers to characterise the origins of intercontinentally-transported biomass burning emissions, ozone and its precursors (Stohl, 1998, 2004; Stohl and Thompson, 1999; Stohl and Trickl, 1999; Stohl *et al*, 1998, 2002a, 2002b, 2003b, 2005; Damoah *et al*, 2004; Forster *et al*, 2001; Huntrieser *et al*, 2005; Trickl *et al*, 2003; Wenig *et al*, 2003);
- On the EMEP website (http://www.emep.int/SR_data/sr.html) one can find data, derived using the EMEP Eulerian model (Simpson *et al*, 2003b), on the contributions of various European countries to pollution by sulphur and nitrogen species, ozone and particulates in neighbouring countries. Similar data are also available for heavy metals and POPs/PBTs (http://www.emep.int/index_pollutants.html). The methodology used is discussed by Tarrasón *et al* (2003);
- Hopke and coworkers have contributed extensively to the application of statistical methods for the quantitative apportionment of air pollutants to their sources (e.g. Hopke *et al*, 1993, 2005; Begum *et al*, 2005). A complete bibliography can be found at <http://people.clarkson.edu/~hopkepk/project1.html>.

5 Potential impacts of Long-Range Transport

The potential impacts of LRT will not be analysed in any depth in the present dossier. The most significant effects will merely be listed, so as to illustrate the concerns that have led to the enacting of national and international regulations aimed at minimising the consequences of LRT. More detailed overviews of impacts can be found elsewhere (e.g. Ritter *et al*, 1995; WHO, 2000; CACAR, 2003b).

5.1 Human Health Impacts

5.1.1 Inhaled pollutants

Certain air pollutants – namely those that are inhaled – can lead to human health effects even without being deposited to the biosphere. The World Health Organization (WHO, 2000) lists four “classical” air pollutants: nitrogen dioxide, ozone, suspended particulate matter (particularly the finer fraction with a diameter less than 2.5 µm) and sulphur dioxide. As discussed above, all these substances may undergo atmospheric LRT. Furthermore, all can contribute to or aggravate respiratory ailments, particularly in sensitive subjects such as asthmatics, children or the elderly (Lübker-Alcama and Krzyzanowski, 1995; WHO, 2000; NARSTO, 2000; IAQAB, 2002; USEPA, 2004; EEA, 2004).

While the concentrations of the pollutants are not necessarily high enough to cause significant harm to human health following LRT over distances of hundreds or thousands of kilometres, there are nevertheless instances when this may occur, especially for ozone and particles. When this is the case, fears are understandably raised that emissions in faraway places may cause the pollution control strategies and efforts of other nations or region to fall short of attaining local air quality objectives.

5.1.2 Ingested pollutants

Humans may also be adversely affected by pollutants arising from distant sources and ingested with drinking water or foodstuffs, for example as a result of LRT through the atmosphere and deposition onto soil, pastures, crops or water bodies, followed – in some cases – by biomagnification up the food chain, ultimately leading to high levels in fish, meat or dairy products.

Examples of particular concern include:

- Toxic metals: After deposition from the atmosphere, inorganic mercury can be converted by environmental microorganisms to the highly potent neurotoxin monomethylmercury, the concentration of which is biomagnified with increasing trophic level up the food chain (ATSDR, 1999a; UNEP, 2002b; CACAR, 2003b). Other toxic metals of special concern are lead and cadmium (ATSDR, 1999b, 1999c; CACAR, 2003b);
- Radionuclides: For instance, the potential global impact of the 1986 nuclear accident in Chernobyl (Ukraine) was assessed by Anspaugh *et al* (1988) and a slightly increased cancer incidence in northern Sweden has recently been linked to this accident (Tondel *et al*, 2004);
- POPs/PBTs: Most of the semi-volatile organic compounds discussed above are lipophilic in nature, especially the PCDDs, PCDFs, PCBs and chlorinated pesticides, including DDT and HCH. They tend to concentrate in the fatty tissues of living organisms and biomagnify from one species to the next up the food chain. Subsequent human dietary exposure to low levels of certain POPs/PBTs may be associated with a wide range of adverse effects, including endocrine disruption, immune dysfunction, neurological deficits, reproductive and developmental anomalies, behavioural abnormalities and carcinogenesis (Ritter *et al*, 1995; CACAR, 2003b).

Much attention has been paid to the impact of bioaccumulating substances on the native populations of the Arctic region, in response to concerns about the elevated concentrations of these contaminants in fish and wildlife species that are important components of the traditional diets of northern aboriginal peoples (CACAR, 2003a, 2003b).

5.1.3 Indirect impacts

“Indirect” effects on human health may result from the impact of persistent chemicals on the environment (to be outlined below). For instance, depletion of the stratospheric ozone layer leads to an enhanced flux of solar ultraviolet radiation at the earth’s surface, which in turn can cause an increased incidence of skin cancer, cataracts, immune disorders, etc. (UNEP, 2003). Climate change, believed to be induced at least partially by anthropogenic chemicals (particularly CO₂, CH₄, N₂O and CFCs), globally dispersed in the atmosphere, can lead to a range of human health impacts, too numerous to even list here (IPCC, 2001a; EEA, 2004; Parmesan and Galbraith, 2004).

5.2 Environmental Impacts

5.2.1 Stratospheric ozone depletion

Volatile organic chlorine and bromine compounds that are emitted in substantial quantities and that have atmospheric persistence greater than about 1 year, contribute to depletion of the stratospheric ozone layer (WMO, 2002). Of particular concern in this respect are the CFCs, the halons, carbon tetrachloride, 1,1,1-trichloroethane and methyl bromide.

5.2.2 Climate forcing

The so-called “anthropogenic greenhouse gases” perturb the radiative balance between the solar visible and ultraviolet radiation reaching the earth’s surface and the infrared energy radiated back to space. It is believed that the resulting “radiative forcing” leads to an increase in the average temperature at ground level and consequently to a whole host of climate-related impacts (IPCC, 2001a, 2001b). The most important greenhouse gases are the long-lived (and hence globally dispersed) compounds CO₂, CH₄, N₂O and the CFCs.

The climate forcing due to aerosols (sulphate, carbonaceous, dust, etc.) is complex and insufficiently characterised. Particles exert both a direct effect by scattering and absorbing solar and infrared radiation and an indirect effect by altering cloud formation processes. Overall, aerosols are probably responsible for a significant cooling effect, although black carbon is an important warming component (IPCC, 2001b, Chapter 5).

The numerous potential impacts of climate change on ecosystems are discussed in various recent reviews (IPCC, 2001a; EEA, 2004; Parmesan and Galbraith, 2004).

5.2.3 Tropospheric ozone formation – Photochemical smog

The formation of ozone and related oxidants in the troposphere by a complex suite of reactions involving CO and short-lived organic species, nitrogen oxides and sunlight, is a well known phenomenon, especially in densely populated areas in summer (NARSTO, 2000; USEPA, 2004, EEA, 2004).

However, in addition to this urban “photochemical smog” or “summer smog”, the background levels of ozone in the remotest areas of the world have been rising inexorably for over a century, on account of LRT of ozone itself or its precursors, so that “global smog” is now becoming a concern (Hough and Derwent, 1990; Fishman *et al*, 1996; Levy *et al*, 1997; Wang and Jacob, 1998; Berntsen *et al*, 2000; IPCC, 2001b, Chapter 6; UNECE, 2003). It has been concluded that intercontinental transport of pollution can at least partially negate the results of local efforts to reduce critical exposure levels of ozone (Langmann *et al*, 2003; UNECE, 2003; Keating *et al*, 2004).

In addition to human health impacts, ozone is believed to contribute to adverse effects on vegetation, such as forest dieback and reduction in crop yields (NARSTO, 2000; USEPA, 2004; Giles, 2005a). In particular, the Mediterranean region has high ozone concentrations in summer (Millán *et al*, 1997), close to levels at which adverse effects on vegetation are observed (Fumagalli *et al*, 2001). Furthermore, ozone is a significant greenhouse gas (Berntsen *et al*, 2000; IPCC, 2001b, Chapter 6).

5.2.4 Acidification / Eutrophication

The increase in the acidity of precipitation, surface waters and soils, arising from anthropogenic emissions of NO and SO₂, which are subsequently transformed into nitric and sulphuric acids, is well documented (Cowling, 1982; Galloway, 1995, 2001; Bouwman *et al*, 2002). In recent years, the atmospheric emission rates of these two gases have

exceeded natural rates by about 8 and 4 times, respectively (Galloway, 2001). Particularly noteworthy effects of increased acidity on ecosystems are mortality of aquatic biota and forest dieback (Nosengo, 2003; Galloway *et al*, 2004; Giles, 2005b). Significant damage to forests in Europe became a high-priority environmental issue around 1980, while lakes and rivers in Scandinavia lost fish populations due to acidification from the 1950s to the 1980s (Cowling, 1982; Matzner and Murach, 1995; UNEP, 2002a; Wright *et al*, 2005). However, SO₂ emissions have passed their peak values in all the major regions of the world and have been reduced very significantly in Europe and North America (Jeffries *et al*, 2003; Stern, 2005; Wright *et al*, 2005), leading to a decrease in acidity of precipitation and surface-water (Ruoho-Airola *et al*, 2004; Fowler *et al*, 2005; Skjelkvåle *et al*, 2005).

It should be noted that ammonia, which also undergoes atmospheric LRT, is another cause for concern. Although it is alkaline, when deposited to terrestrial ecosystems it can be converted to nitrate and exert an acidifying effect on soils and groundwaters. In some areas, soil acidification from nitrification of ammonia deposited from the atmosphere may be comparable to that from deposition of nitric acid (Galloway, 1995).

Non-biological impacts of atmospheric acidity are corrosion of metals and stone, the latter involving deterioration of mankind's long-term cultural heritage (Kucera and Fitz, 1995).

Cowling (1982) provides a detailed historical review on acid precipitation, from the 17th century to 1982, documenting in particular the rise of awareness in the 1960s and 1970s of the environmental consequences of this phenomenon.

In addition to the impact of acidity as such, nitrate and other forms of "fixed" (or "reactive") nitrogen can lead to the fertilisation and ultimately eutrophication of open-ocean, coastal, freshwater and terrestrial ecosystems, as well as to the contamination of groundwater. While nutrients play an essential role in aquatic environments, problems arise when inputs become excessive and/or the ratio between nutrients is substantially changed. Typical examples of such problems are increased algal growth, changes in the biological community structure and biodiversity, including the occurrence of harmful algae, oxygen depletion and mass mortality of benthic organisms and fish. For instance, occasional "red tides" or coastal algal blooms are observed that may be devastating in their impact (Kahru *et al*, 2004; Cloern *et al*, 2005) and the Baltic Sea is suffering from severe eutrophication (Rönnerberg and Bonsdorff, 2004). The LRT impact of eutrophying pollutants may result either from atmospheric deposition of nitrate or ammonia, or from riverine transport of dissolved nitrogen species (Galloway *et al*, 1995; Anderson *et al*, 2002; Prepas and Charette, 2003; Bouwman *et al*, 2002; Bouwman *et al*, 2005). Increasing reactive nitrogen in the environment and the resulting eutrophication is regarded by certain authors as one of the major challenges of global change (Nosengo, 2003; Galloway *et al*, 2004; Giles, 2005b).

5.2.5 *Visibility impairment*

Various types of atmospheric aerosols lead to visibility impairment. Classic examples involving LRT include the Arctic haze and the "brown cloud" that frequently extends over broad areas of Asia (ASEAN, 2001; Wooster and Strub, 2002; UNEP-C⁴, 2002; Park *et al*, 2003; CACAR, 2003a; Langmann and Heil, 2004; Benkovitz *et al*, 2004; Cyranoski and Fuyuno, 2005).

5.2.6 *Mercury*

The potential toxicological effects of mercury on wildlife (central nervous system, kidneys, reproduction, etc.) and ecosystems have been reviewed by UNEP (2002b).

5.2.7 *Persistent organic pollutants*

Experimentally, POPs/PBTs have been associated with significant environmental impacts in a wide range of species at virtually all trophic levels. For certain POPs/PBTs, there is some experimental evidence that low-level exposures may be associated with chronic non-lethal effects including endocrine disruption, immunotoxicity, impairment of reproductive performance and carcinogenicity (Ritter *et al*, 1995; AMAP, 2004). A discussion of the validity of these findings is beyond the scope of this dossier.

5.2.8 Interactions

The linkages between the impacts of “traditional” air pollutants and of those responsible for climate change have been reviewed by EEA (2004).

5.3 Evolution of the perceived “spatial scale” of air pollution

Keating *et al* (2004) point out that, while certain environmental and human health impacts have always been considered to be more or less global in scope (nuclear fallout, stratospheric ozone depletion and climate forcing), in other cases the perception of the “spatial scale” has evolved over the past decades. Thus, for photochemical smog, acid rain and aerosol particles, a gradual reassessment of the issues has occurred over time, with the focus shifting from the local or regional scale to a more global perspective.

Keating *et al* (2004) describe the various pressures leading to a “tightening vice” of air pollution management. On the one hand, local emissions of the most acutely harmful air pollutants have been abated fairly successfully – at least in the developed countries – and further reductions are more and more difficult and costly to achieve. On the other hand, local air quality standards are becoming more stringent, while at the same time emissions in the developing world are rising and leading to increases in the regional and/or global background pollutant concentrations.

Consequently, there is an increasing emphasis on pollutants prone to LRT, as regulators look to “upwind” jurisdictions to control their share of emissions.

6 International Regulatory and Monitoring Agreements

In view of the observed LRT and potential impacts of a wide range of substances, as discussed above, a number of international agreements have been concluded in order to address and alleviate the underlying problems. Essentially, this is to be achieved by prescribing reductions in emissions, restrictions on use, or ultimate phase-out.

6.1 Agreements that Explicitly Address Transboundary Pollution or Long-Range Transport

6.1.1 UNECE LRTAP Convention

Broad international cooperation to combat the impacts that pollutants emitted from one nation have on its neighbours or more distant regions began in 1979, when the United Nations Economic Commission for Europe (UNECE) concluded its framework Convention on Long-range Transboundary Air Pollution (<http://www.unece.org/env/lrtap/>). This agreement now has 49 parties, including not only “European” countries, interpreted broadly, but also the United States and Canada.

Although the LRTAP Convention was aimed initially at responding to the challenge posed by acid rain, a total of eight protocols have now been negotiated, identifying specific measures to be taken by the parties for a range of air pollutants:

- The 1984 Geneva Protocol on Long-term Financing of the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP);
- The 1985 Helsinki Protocol on the Reduction of Sulphur Emissions or their Transboundary Fluxes by at least 30 per cent;
- The 1988 Sophia Protocol concerning the Control of Nitrogen Oxides or their Transboundary Fluxes;
- The 1991 Geneva Protocol concerning the Control of Emissions of Volatile Organic Compounds or their Transboundary Fluxes (targeting the most reactive volatile compounds);
- The 1994 Oslo Protocol on Further Reduction of Sulphur Emissions;
- The 1998 Aarhus Protocol on Heavy Metals (targeting mercury, cadmium and lead);
- The 1998 Aarhus Protocol on Persistent Organic Pollutants (POPs), targeting aldrin, chlordane, chlordecone, DDT, dieldrin, endrin, HCHs, heptachlor, hexabromobiphenyl, hexachlorobenzene, mirex, PAHs, PCBs, PCDDs, PCDFs, pentachlorophenol and toxaphene, with provisions for addition of further substances in the future;
- The 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (targeting sulphur, nitrogen oxides, ammonia and reactive VOCs).

All of these protocols have been signed and ratified by sub-sets of the parties to the 1979 Convention and all have entered into force.

The LRTAP Convention is backed up by EMEP (<http://www.emep.int/>), a co-operative programme for monitoring and evaluation of the LRT of air pollutants in Europe.

6.1.2 UNEP POPs – Stockholm Convention

An important milestone in international cooperation on global dispersion and impact of pollutants was the conclusion in 2001 of the United Nations Environment Programme’s Stockholm Convention on Persistent Organic Pollutants (<http://www.pops.int/>). This agreement, whose geographical representation is much broader than that of the UNECE POPs protocol – since it includes many non-European and developing states – entered into force in 2004 and currently has 112 parties. It targets the following 12 compounds or families of compounds: aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, PCBs, PCDDs, PCDFs and toxaphene. There are provisions for the addition of further substances.

6.1.3 U.S. - Canada Air Quality Agreement

The 1991 "Agreement Between the Government of the United States of America and the Government of Canada on Air Quality" (<http://www.epa.gov/airmarkets/usca/agreement.html>) was negotiated mainly to reduce emissions of SO₂ and NO_x and hence the transboundary acid deposition caused by these gases. However, an annex to the agreement also provides for the monitoring of other air pollutants of concern.

6.1.4 The Great Lakes Binational Toxics Strategy

The Canada - United States "Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes" (<http://www.epa.gov/glnpo/p2/bns.html>), was approved in 1997. It is based on the "Revised Great Lakes Water Quality Agreement" of 1978 (<http://www.ijc.org/php/publications/pdf/ID609.pdf>) and covers not only a range of organochlorines, PAHs and other POPs/PBTs, but also certain compounds of mercury, cadmium, lead and tin. Although the focus of the treaty is on pollution of the Great Lakes by emissions from within the United States and Canada, LRT from worldwide sources is also considered explicitly.

6.1.5 The International Joint Commission (U.S. - Canada)

The International Joint Commission (http://www.ijc.org/en/home/main_accueil.htm) assists the United States and Canada in the protection of the transboundary environment, including the implementation of the "Great Lakes Water Quality Agreement" and the improvement of transboundary air quality.

6.1.6 North American Agreement on Environmental Cooperation

In 1993, the NAFTA countries Canada, Mexico and the United States signed the North American Agreement on Environmental Cooperation (NAAEC). A draft "North American Agreement on Transboundary Environmental Impact Assessment" was published in 1997 and the current "Agenda for Action" of the North American Commission for Environmental Cooperation (NACEC, <http://www.cec.org>) includes provisions for addressing air pollution problems within transboundary airsheds: (http://www.cec.org/pubs_info_resources/law_treat_agree/pbl.cfm?varlan=english) (http://www.cec.org/files/pdf/POLLUTANTS/311-03-05_en.pdf).

NACEC's "Sound Management of Chemicals" project aims, *inter alia*, at identifying priority chemical pollution issues of regional concern. Protocols on both POPs/PBTs and metals (Hg, Cd, Pb) are being considered: (http://www.cec.org/programs_projects/pollutants_health/project/index.cfm?projectID=25&varlan=english).

6.1.7 ASEAN Agreement

In 1994, the 10 Southeast Asian nations belonging to the ASEAN partnership agreed to the formulation of a "Cooperation Plan on Transboundary Pollution" (<http://www.aseansec.org/8938.htm>). Although not restricted to atmospheric pollution, a major motivation of this agreement was clearly combating the haze from forest fires and biomass burning that regularly affects the whole region (ASEAN, 2001). Indeed, a specific "ASEAN Agreement on Transboundary Haze Pollution" was signed in 2002 (<http://www.fire.uni-freiburg.de/media/2003/WSSD-ASEAN-Agreement.pdf>).

6.1.8 East Asia Network

The "Acid Deposition Monitoring Network in East Asia" (EANET, <http://www.adorc.gr.jp/>) was initiated in 1998 by 12 East Asian Nations, including Japan, China, Korea and Russia. This cooperation covers only scientific activities on acid deposition and atmospheric oxidants, without any regulatory prerogatives.

6.1.9 Malé Declaration

Eight South Asian governments approved the 1998 "Malé Declaration on Control and Prevention of Air Pollution and its Likely Transboundary Effects for South Asia". The stated aim of this agreement is to achieve intergovernmental cooperation to address the increasing threat of transboundary air pollution and consequential impacts due to concentrations of pollutant gases and acid deposition on human health, ecosystem function and corrosion of materials (http://www.ricap.unep.org/issues/air/maledec/aw_md.cfm).

6.1.10 Harare Resolution

Seven southern African nations endorsed the 1998 “Harare Resolution on the Prevention and Control of Regional Air Pollution in Southern Africa and its likely Transboundary Effects” (<http://www.york.ac.uk/inst/sei/rapidc2/apina/apina-resolution.html>).

6.1.11 Arctic Council activities

The “Plan to Eliminate Pollution of the Arctic”, approved in 2000 by the Arctic Council, i.e. an intergovernmental forum of the nations bordering the Arctic, was created to address pollution sources identified through the Arctic Monitoring and Assessment Programme, established in 1991 (Arctic Council, 2001; AMAP, 2002, 2004; <http://www.arctic-council.org/en/main/infopage/5/#amap>). The transboundary nature of such pollution is self-evident. First-phase priority will be given to POPs/PBTs, heavy metals, radioactivity and ozone-depleting substances.

The “Programme for the Protection of the Arctic Marine Environment (PAME)”, established in 1983, is another initiative of the Arctic Council involving transboundary pollution (<http://www.pame.is/>).

6.1.12 OSPAR Convention

The 1992 Oslo and Paris (OSPAR) “Convention for the Protection of the Marine Environment of the North-East Atlantic” (<http://www.ospar.org/>) entered into force in 1998. Its contracting parties are 15 European countries and the European Community. OSPAR aims to prevent pollution of the North-East Atlantic by continuously reducing discharges, emissions and losses of hazardous substances (identified by specific “persistence, bioaccumulation and toxicity” criteria), with the ultimate aim of achieving concentrations in the marine environment near background values for naturally-occurring substances or close to zero for man-made substances. The prevention of pollution from land-based sources, as well as offshore ones, dumping and incineration are specifically cited in the Convention. Atmospheric, riverine and anthropogenic LRT is therefore a concern.

6.1.13 Helsinki Convention

The 1992 Helsinki “Convention on the Protection of the Marine Environment of the Baltic Sea Area” (HELCOM: <http://www.helcom.fi/helcom.html>) entered into force in 2000. The contracting parties are seven nations bordering the Baltic, and the European Community. The aims and focus of the Convention are similar to those of OSPAR, but transposed to the Baltic.

6.1.14 Barcelona Convention

The 1976 Barcelona “Convention for the Protection of the Marine Environment and the Coastal Region of the Mediterranean” (<http://www.unepmap.org/home.asp>) entered into force in 1978. The contracting parties are 21 states bordering the Mediterranean Sea, and the European Community. Protocols to the agreement relate to protection against pollution of the Mediterranean from land-based sources, dumping of hazardous wastes, etc..

6.1.15 UNEP Regional Seas Programme

The “Global Programme of Action for the Protection of the Marine Environment from Land-Based Activities” (<http://www.gpa.unep.org/default.htm>) was initiated by UNEP in 1974 and currently includes 13 regions, with 140 participating coastal states. The Barcelona Convention is the legal instrument for UNEP’s “Mediterranean Action Plan” (<http://www.unepmap.org/>), which is one of the components of the Regional Seas programme. Similarly, the legal framework for the “Caribbean Action Plan” is the Cartagena Convention, adopted in 1983 (<http://www.cep.unep.org/law/cartnut.html>).

6.1.16 Basel Convention

UNEP’s “Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal” (<http://www.basel.int/>) was adopted in Basel in 1989. It entered into force in 1992 and counts most of the world’s nations as parties. This convention aims at reducing cross-border movements of hazardous wastes. It therefore addresses certain problems posed by “anthropogenic” LRT.

6.2 Agreements in which Long-Range Transport is Implicitly Included

In addition to the agreements listed above, in which transboundary pollution is addressed as a specific concern, there are other major international treaties that do not explicitly mention LRT, but whose focus is nevertheless on environmental and human health impacts of a global nature, caused by the release of substances that become broadly dispersed in the environment.

Particularly noteworthy in this respect are the international conventions addressing stratospheric ozone depletion and climate change. The compounds currently regulated under these agreements are all volatile substances having atmospheric lifetimes ranging from months to millennia. They are thus highly prone to LRT and become distributed throughout the atmosphere on a hemispheric or even global scale.

6.2.1 *Montreal Protocol*

UNEP's 1987 "Montreal Protocol [to the 1985 Vienna Convention] on Substances that Deplete the Ozone Layer", together with the various Amendments and Adjustments agreed since its entry into force in 1989 (<http://www.unep.org/ozone>), aims at reducing emissions of ozone-depleting substances and ultimately phasing them out. The target compounds are CFCs, HCFCs, halons, carbon tetrachloride, 1,1,1-trichloroethane and methyl bromide.

6.2.2 *Kyoto Protocol*

UNEP's 1997 "Kyoto Protocol to the United Nations Framework Convention on Climate Change" (http://unfccc.int/essential_background/kyoto_protocol/items/2830.php) has the objective of reducing emissions of the main greenhouse gases: CO₂, CH₄, N₂O, HFCs, PFCs and SF₆. Despite their significant contribution to global warming, CFCs are not included, since they are scheduled for phase-out under the Montreal Protocol.

7 Regulatory Criteria Expressing the Propensity of a Substance to Undergo Long-Range Transport

Certain of the international treaties discussed in the previous chapter, as well as some national or regional regulations, include a criterion for expressing the ability of a substance to undergo LRT. This characteristic is generally one item of a set of criteria used to determine whether or not a compound should be classified as a POP/PBT. These sets of criteria will be used for adding new compounds to the lists of substances covered by the respective agreements or regulations. However, some form of expert judgement may also be required, in addition to strict compliance with the numerical “bright-line” criteria, before a new compound is listed.

Not all definitions of POPs/PBTs include a specific LRT criterion, although many do have a criterion for persistence in the atmosphere, which effectively determines the potential for atmospheric LRT.

The main POP/PBT criteria have been summarised by Euro Chlor (2003).

7.1 Monitoring evidence

In the case of the UNECE-LRTAP Aarhus Protocol on POPs, the procedure for addition of new substances includes submitting a “risk profile”, based on criteria defined by UNECE Executive Body Decision 1998/2

(<http://www.unece.org/env/documents/2000/ece/eb/ece%20eb%20air.60.e.pdf>). The potential for long-range transboundary atmospheric transport is defined by certain physico-chemical properties of the substance (to be discussed below) or, alternatively, by “monitoring data showing that the substance is found in remote regions”.

In the case of the UNEP Stockholm Convention on POPs, the screening criteria for additional compounds are listed in its Annex D (http://www.pops.int/documents/convtext/convtext_en.pdf). Information is to be provided on “measured levels of the chemical in locations distant from the sources of its release that are of potential concern”. Additionally, the criterion “monitoring data showing that long-range environmental transport of the chemical, with the potential for transfer to a receiving environment, may have occurred via air, water or migratory species” is listed as a possible alternative to consideration of certain environmental fate properties or model results (to be discussed below).

NACEC’s draft “Sound Management of Chemicals Process (SMOC) for Identifying Candidate Substances for Regional Action” (http://www.cec.org/programs_projects/pollutants_health/smoc/criter.cfm?varlan=english) adopts similar language, since it provides for examining “monitoring evidence of transboundary transport for metals or POPs/PBTs (e.g. appearance in biota)”.

The Canadian Toxic Substances Management Policy (TSMP) includes persistence in any one of four media as one of the criteria to be fulfilled for scheduling chemicals for virtual elimination. The regulation states that “a substance may be considered as persistent in air if it is shown to be subject to atmospheric transport to remote regions such as the Arctic” (<http://www.ec.gc.ca/toxics/TSMP/en/track1.cfm>).

It should be borne in mind that monitoring evidence as a measure of LRT can only be used *a posteriori*, once the substance has been released to the environment and not prior to its commercialisation.

7.2 Modelling evidence

The UNEP POPs Convention adopts, as evidence for LRT potential and as an alternative to monitoring data, “environmental fate properties and/or model results that demonstrate that the chemical has a potential for long-range environmental transport through air, water or migratory species, with the potential for transfer to a receiving environment in locations distant from the sources of its release”. There is an additional proviso in this respect that “for a chemical that migrates significantly through the air, its half-life should be greater than two days”.

The UNECE-LRTAP POPs Protocol defines a procedure for reviewing “risk profiles” submitted with a view to adding new substances. This procedure includes an evaluation of “the monitoring or equivalent scientific information suggesting long-range transboundary atmospheric transport”. It is likely that modelling evidence would be deemed “equivalent” to monitoring.

7.3 Vapour pressure

The UNECE-LRTAP POPs Protocol includes, as a criterion for atmospheric LRT potential, “evidence that the substance has a vapour pressure below 1,000 Pa and an atmospheric half-life greater than 2 days”. The vapour pressure criterion warrants some explanation. Clearly, high volatility would be no impediment to atmospheric LRT: indeed, the opposite is the case. Some clue as to the motivation behind the volatility criterion is given by a background document to the UNECE criteria (AEAT, 1995), which states that “substances with vapour pressure > 1000 Pa are considered so volatile that they will remain predominantly in the atmosphere and not transfer to the condensed phase”. This is only partially true: for instance, some substances are very volatile, but also highly soluble in water, so they would partition to aqueous compartments. In any case, the 1000 Pa upper limit does not represent a low enough volatility for substantial uptake of the substance to particles in the atmosphere, which would require a vapour pressure about 5 orders of magnitude lower (Bidleman, 1988). The fraction of the substance that partitions to other “condensed” phases (water, soil, sediment, biota) depends not only on volatility, but also on the affinity of the substance for these phases (hydrophilicity, lipophilicity). So it would seem that the 1000 Pa cut-off is just a crude demarcation that enables one to avoid classifying as POPs a large number of very volatile compounds (including those targeted by the Montreal and Kyoto Protocols), without having to go through an assessment of their toxicity or tendency to bioaccumulate.

The NACEC-SMOC draft includes this same “vapour pressure < 1000 Pa” criterion, also with the proviso that air persistence is > 2 days.

Although some volatility is obviously necessary for atmospheric LRT, compounds with extremely low vapour pressures have been found in remote regions far from their presumed sources. This is the case, for example, for a number of currently used pesticides, having vapour pressures ranging from 10^{-8} to 5×10^{-2} Pa, found in an ice core from the Norwegian Arctic (Hermanson *et al*, 2005).

7.4 Atmospheric half-life

An atmospheric half-life greater than 2 days is a common criterion, either for LRT as such or for persistence. This numerical value is included in the following treaties and regulations: UNECE-LRTAP POPs Protocol, UNEP POPs Convention, NACEC-SMOC and Canada-TSMP. The scientific basis for this value has been discussed by Bidleman (1997) and Rodan *et al* (1999). As has been mentioned above, a 2-day half-life roughly corresponds to the lower limit for intercontinental atmospheric transport, except under exceptional meteorological circumstances.

7.5 Half-life in water

Many treaties and regulations do not list any specific criteria for LRT or for persistence in the atmosphere, but do assess chemicals on the basis of their persistence in other media. These include marine or fresh water, which is of course relevant to riverine and oceanic LRT. Half-lives in water greater than thresholds between 40 days and 6 months are

generally adopted as a criterion for persistence in water (Rodan *et al*, 1999; Euro Chlor, 2003).

8 Summary and Conclusions

Long-Range Transport (LRT) is a phenomenon which encompasses many classes of natural and anthropogenic substances and affects every point of the globe. It is, nevertheless, often presented and perceived merely as a process by which semi-volatile organic compounds are emitted at tropical or temperate latitudes, migrate northwards through the atmosphere in a series of volatilisation-recondensation cycles or "hops", and finally accumulate in the Arctic and Antarctic food chain, to the detriment of the indigenous populations. While this image is not totally incorrect, the present dossier amply demonstrates that it is overly simplistic.

The atmosphere being the most mobile of the environmental media, air is a major vector for LRT. However, it is not the only one: for non-volatile, water-soluble substances, transport along rivers and through the oceans is preponderant. Furthermore, in specific cases, migratory animal species, drifting ice and anthropogenic transport for commercial or other reasons can all play a role in the worldwide environmental dispersion of chemicals.

Intercontinental atmospheric transport generally occurs on a time-scale of about 3-30 days. Pollutants lofted into the free troposphere by deep convection, mountain-slope forcing or frontal systems will typically travel further and faster than those that are simply advected horizontally in the atmospheric boundary layer. The time-scale for dissolved species to be transported by ocean currents from northern mid-latitudes to the Arctic is several years.

Volatile pollutants with atmospheric lifetimes of years or longer experience no impediment to LRT once they are emitted to air. Whatever the location and timing of their release, their concentrations become practically uniform throughout the atmosphere, so the emission of a given amount anywhere on Earth makes the same contribution to global pollution. This is the case, for example, for the major greenhouse gases carbon dioxide, methane and nitrous oxide, as well as for the ozone-depleting chlorofluorocarbons and halons. For air pollutants with lifetimes of several months, concentrations become fairly uniform within the hemisphere of emission, but not in the opposite hemisphere, since crossing the equatorial "barrier" takes about a year. For shorter-lived pollutants with atmospheric lifetimes of days or weeks, the propensity of a substance to undergo atmospheric LRT depends on its intrinsic physico-chemical properties (reactivity, volatility and water solubility), as well as on its mode of entry into the environment, the location and time of its emission and the prevailing meteorological conditions.

Atmospheric transboundary and intercontinental LRT is well documented for many classes of substances. While some of these are purely or primarily anthropogenic (e.g. pesticides, PCBs, lead, and radioactive fallout from nuclear testing and accidents), others have partly natural and partly man-made sources. Examples of such substances of mixed origin are carbonaceous particles, carbon monoxide, nitric oxide and other combustion by-products; ozone arising from atmospheric reactions of the afore-mentioned; sulphur dioxide and sulphate aerosols; mercury; and a range of volatile organics including some halogenated species, such as methyl chloride, methyl bromide, chloroform and bromoform. Finally, a number of substances of exclusively natural origin are also well known to undergo intercontinental LRT. Examples are desert sand, soil dust, volcanic ash, sea salt, pollen, pathogenic microbes and natural radionuclides (e.g. ^{210}Pb resulting from the decay of radon).

Contributions to understanding the major atmospheric LRT pathways have been made by analysing samples taken at ground level, as well as from balloons and aircraft, with back-up from satellite observations and modelling studies.

The winds blowing from the west at mid-latitudes exert a major influence on intercontinental atmospheric LRT, leading to certain preferential pathways. There is an increasing awareness of transport of pollution from Asia to North America. The substances involved include ozone and its precursors, suspended particles, sulphur dioxide and sulphate aerosols, pesticides and mercury. In turn, Europe is affected by pollutants from North

America, while Asia is impacted by European emissions. In the southern hemisphere, there is also evidence for eastward transport of pollution, from southern Africa to Australia, New Zealand and the islands of the southern Pacific. Towards the equator, the easterly trade winds tend to govern atmospheric circulation, as illustrated by the transport of sand from North African deserts to the central part of the American continent. In the polar regions too, the air flow is mainly from the east. However, the general pattern of westerly or easterly circulation is disrupted over broad areas of the globe, so that atmospheric LRT also occurs in other directions, often with a seasonal dependence. A few illustrative examples are: transport of Saharan dust northwards to Europe in spring and summer; inflow of "haze" (mainly due to sulphur species) into the Arctic from Eurasia in winter; the contamination of Sweden and Finland by fallout from the nuclear accident at Chernobyl in Ukraine; and the presence in the "pristine" polar ice-caps of pesticides and heavy metals (including Pb from metallurgical operations carried out several millennia ago).

Air quality at a given location can be influenced by emissions from another continent either through an increase in the more or less ubiquitous hemispheric background levels of pollution, or through discrete episodic flows of enhanced pollutant levels.

A broad range of numerical models has been developed and applied for predicting how chemicals are distributed in the environment and for assessing their overall persistence and long-range transport potential (LRTP). These models range in complexity from simple "evaluative" multimedia box models to three-dimensional, georeferenced general circulation chemistry-transport models with high spatial resolution. Several comparative studies have led to the conclusion that most models predict similar rankings of LRTP values for sets of chemicals encompassing widely varying physico-chemical properties. Most models account for the "grasshopper effect" by which semi-volatile chemicals undergo repeated cycles of volatilisation and partitioning to condensed phases, while some models even allow for the calculation of the number of "hops" taken by a chemical on its journey from warmer to colder latitudes. Certain models have been developed for the express purpose of exploring source-receptor relationships, i.e. to answer questions such as "Where does pollution at this location come from?" or "Which downwind areas will be affected by emissions from regions of production?"

It should be emphasised that LRT is not in itself a concern, but only a trigger for potential concern, which will be warranted if the pollutant is demonstrated or predicted to have an impact at a receptor site. Such an impact may affect the atmosphere itself (e.g. ozone depletion, global warming or photochemical smog formation) or may cause an adverse effect on human health through inhalation, in which case the pollutant will be harmful even without being deposited to the Earth's surface. In many instances, however, deposition followed by uptake into living organisms, possibly with accumulation through the food chain, is a prerequisite for damage to the environment or human health. Each chemical needs to be evaluated in this respect on a case-by-case basis, with the potential for LRT being assessed in conjunction with potentially adverse properties, such as persistence, bioaccumulation, toxicity and the capacity to affect the atmosphere.

Numerous impacts may be caused by substances transported to locations remote from their emission sources. Human health may be adversely affected by inhalation of airborne pollutants (e.g. nitrogen dioxide, ozone, suspended particles, or sulphur dioxide, all of which contribute to respiratory ailments) or by ingestion of deposited substances with food or drinking water (e.g. heavy metals, radionuclides or POPs/PBTs, each substance having its own diverse toxicological end-points). The environment itself is also vulnerable to certain long-range transported chemicals. Apart from the atmospheric impacts noted above (namely ozone depletion, climate warming and photochemical smog, all of which can be harmful to both human health and ecosystems), major concerns arise from transboundary pollution by sulphur dioxide, nitrogen oxides and ammonia, which are converted into acids that cause damage to forests, lakes and rivers, as well as to materials of construction. In addition to their role in acidification of the environment, nitrogen oxides also lead to eutrophication of freshwater bodies and coastal zones, subsequent to LRT of NO_x through the atmosphere or of nitrate with groundwater and rivers. Visibility impairment of broad areas of the Earth is another consequence of atmospheric LRT. Noteworthy examples are the Arctic haze and the huge Asian "brown cloud". For certain POPs/PBTs, there is some evidence that low-level exposures may be associated with chronic non-lethal effects

including endocrine disruption, immunotoxicity, dermal effects, impairment of reproductive performance and carcinogenicity.

Broad international cooperation to combat the impacts that pollutants emitted from one nation have on its neighbours or more distant regions began in 1979, when the United Nations Economic Commission for Europe (UNECE) concluded its framework Convention on Long-range Transboundary Air Pollution. This agreement, whose parties include the United States and Canada as well as European nations, was aimed initially at responding to the challenge posed by acid rain, but a total of eight protocols have now been negotiated. These cover, amongst others, tropospheric ozone precursors, heavy metals, POPs/PBTs and substances responsible for eutrophication. Another important milestone in international cooperation on global dispersion and impact of pollutants was the conclusion in 2001 of the United Nations Environment Programme's Stockholm Convention on Persistent Organic Pollutants, whose geographical representation is much broader than that of the UNECE POPs protocol – since it includes many non-European and developing states. In addition to these two conventions that fall under the aegis of the United Nations, there are many other regional agreements whose principal or ancillary objective is to protect various territories or seas from transboundary pollution. Such agreements cover: the NAFTA countries (USA, Canada and Mexico); the North American Great Lakes; various groupings of Asian countries; an alliance of southern African nations; the Arctic; the North-East Atlantic; the Baltic; the Mediterranean; the Caribbean; and a number of other “regional seas”.

Many of the major international agreements on harmful substances define criteria expressing the propensity of a compound to undergo LRT. This information can be used in conjunction with other properties (persistence in various environmental media, tendency to bioaccumulate, toxicity, ozone depleting and global warming potentials, etc.) to determine whether or not to schedule the chemical concerned for regulatory action. A common metric for LRT is atmospheric persistence, characterised by a half-life greater than 2 days (i.e. a lifetime > 3 days), which – as mentioned above – is the approximate lower limit for intercontinental atmospheric transport. A secondary criterion in several conventions is “monitoring evidence in remote regions” (such as the Arctic). This measure of LRT can, however, only be used *a posteriori*, once the substance has been released to the environment and not prior to its commercialisation.

While certain environmental and human health impacts of chemicals have always been considered to be more or less global in scope (nuclear fallout, stratospheric ozone depletion and climate forcing), in other cases the perception of the “spatial scale” has evolved over the past decades. Thus, for photochemical smog, acid rain and aerosol particles, a gradual reassessment of the issues has occurred over time, with the focus shifting from the local or regional scale to a more global perspective. Indeed, local emissions of the most acutely harmful air pollutants have been abated fairly successfully – at least in the developed countries. Further reductions are more and more difficult and costly to achieve. On the other hand, local air quality standards are becoming more stringent, while at the same time emissions in the developing world are rising and leading to increases in the regional and/or global background pollutant concentrations – tropospheric ozone being a noteworthy example. Consequently, there is an increasing emphasis on pollutants prone to LRT, as national regulators become only too aware of their diminishing scope for effective action and look to “upwind” jurisdictions to control their share of emissions. Indeed, fears are understandably raised that intercontinental transport of pollution will at least partially negate the results of local efforts to reduce critical exposure levels.

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