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Modelled atmospheric contribution to nitrogen eutrophication in the English Channel and the southern North Sea

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HIGHLIGHTS

- Weather data, emissions data and Lagrangian computations form the numerical model.
- Deposition maps in a target geographical region are produced and compared.
- Individual polluting industrial or transport sectors can be traced.
- Atmospheric deposition of nitrogen eutrophicants increases in wet weather.
- Heaviest atmospheric deposition adds to high riverine input in some coastal regions.

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ABSTRACT

Eutrophication of the coastal waters results in algal blooms which may be harmful to the marine ecosystem and coastal economy. The main sources of nutrients are the rivers but an unquantified amount of nitrogen is also transported from ground sources via the atmosphere and deposited to the sea directly by rain and turbulent diffusion. A Lagrangian Particle Dispersion (LPD) model based on the open source code FLEXPART (http://flexpart.eu) is described that quantifies the dissolved nitrogen coming from the air in the English Channel and Southern North Sea (the '2Seas' geographical region). The model uses meteorological records, emissions data and LPD computations to simulate the motion and deposition of nitrogen compounds. The emission sources contributing to the deposition are individually identified, and calculated concentrations are compared with ground measurements in selected locations. The highest calculated atmospheric depositions to the sea in the considered region are found to be along the Belgium–Netherlands coast.

1. Introduction

Nutrient enrichment of estuaries and coastal waters by human activities causes phytoplankton and algae to grow more than it would do otherwise (e.g. Peierls et al., 1991; Anderson et al., 2002). Provided adequate light is available, phosphorus and nitrogen are the nutrients that limit phytoplankton growth in aquatic systems, and primary production in estuaries and coastal waters is thought to be limited by nitrogen availability (Anderson et al., 2002). In contrast, freshwater phytoplankton tends to be limited by phosphorus availability, although the extent and severity of nitrogen limitation remains open to question (Hecky and Kilham, 1988; Boynton et al., 1982; Nedwell et al., 2002) and in situations where light penetration is poor as in turbid estuarine and coastal waters, even high nutrient concentrations may not lead to significant algal growth. As a societal problem, eutrophication and consequent algal overgrowth has several undesirable socioeconomic and environmental consequences. The additional growth does not enter the marine food chain and by decaying depletes the water of oxygen and thus causes harm to marine life. Another side effect of the decay is unsightly foam on the beaches, which affects tourism and dependant commercial activities.

Groundwater transport constitutes the main source of eutrophicants and therefore the main target of regulative corrective measures (Anderson et al., 2002). Nevertheless, atmospheric deposition is also significant adding from 300 to 1000 mg m^{-2} yr^{-1} to coastal waters in biologically active forms NO_x, NH_3/NH_4^+, and in dissolved organic nitrogen (Paerl, 1997). Atmospheric tracer-based model results show that atmospheric deposition accounts for 6% of

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the external nitrogen inputs in the North Sea (Troost et al., 2013). This percentage however is shown to vary strongly with region. For example, in the OSPAR area NL-O2 (OSPAR, 2013), 16% of the total nitrogen originates from atmospheric deposition. Furthermore, model results show that primary production rates are disproportionately affected by atmospheric deposition, possibly due to a change in the carbon-to-nitrogen ratio (Troost et al., 2013). Budgeting approaches (Spokes and Jickells, 2005) suggest that the atmosphere can in some situations provide enough nitrogen to produce a large increase in phytoplankton growth. The modelled regional and temporal variation reflects the highly episodic nature of atmospheric deposition and the strong gradients away from source regions.

A parallel study in the Baltic Sea shows that atmospheric deposition, primarily from burning fossil fuels (land based and shipping), accounts for 25% of nitrogen input (WRI, 2014). Even more significant atmospheric contributions were found in Chesapeake Bay, U.S (up to 30% of all nitrogen inputs) and in some other areas in the U.S. North Atlantic, where atmospheric deposition of nitrogen can exceed riverine nitrogen inputs to coastal areas (Spokes and Jickells, 2005). It is evident from these observations, that the atmospheric contribution is an essential part of any inventory of nutrients leading to algal growth.

As stated above, the nutrients phytoplankton species need are inorganic compounds of nitrogen, phosphorus and silicon. Of these only nitrogen-bearing compounds such as nitrogen oxides and ammonia, being gases, can be airborne in significant quantities. While in some cases of blooming phytoplankton the availability of phosphorus may be the growth-limiting factor (Ly et al., 2014), nitrogen may still play a part for some species. So a model of the atmospheric input of eutrophicants is a valuable tool in the study, prediction and prevention of harmful algal blooms.

This paper is an illustration of how the three main components of such a model – weather data, emissions data and computer simulations – can be combined into a working tool for quantitative estimates of the atmospheric inputs.

2. Lagrangian transport computer model

Emitted nitrogen-containing gases are carried and dispersed by the wind before being deposited to the ground or sea. A publically available, open source software package FLEXPART (Stohl et al., 2005) implementing the Lagrangian Particle Dispersion method is used in this study. It can simulate the movement of pollutants in the atmosphere and includes also algorithms for determining the rates of their deposition onto various surfaces (e.g. Plainiotis et al., 2005a, 2005b, 2010). In the model, each of the traced 'particles' is assumed to be carrying a certain quantity of the investigated substance. Concentrations are calculated after dispersion by atmospheric turbulence is taken into account. From the concentrations, using specific deposition properties of each traced gas, deposited quantities are calculated on a user-prescribed grid.

2.1. FLEXPART equations

2.1.1. Particle trajectory calculations

The trajectory equation (Stohl et al., 1998)

$$\frac{\mathrm{d}\boldsymbol{X}}{\mathrm{d}t} = \boldsymbol{v}[\boldsymbol{X}(t)]$$

with $\boldsymbol{X}$ the position vector, $\boldsymbol{v}$ the particle velocity, t the time and Δt the time step is integrated using the "zero acceleration" scheme

$$\boldsymbol{X}(t+\Delta t) = \boldsymbol{X}(t) + \boldsymbol{v}(\boldsymbol{X},t)\Delta t.$$

The wind vector $\boldsymbol{v} = \overline{\boldsymbol{v}} + \boldsymbol{v}_t + \boldsymbol{v}_m$ is composed of the grid-scale wind $\boldsymbol{v}$, the turbulent wind fluctuations $\boldsymbol{v}_\mathrm{t}$ and the mesoscale wind fluctuations $\boldsymbol{v}_\mathrm{m}$ (Stohl et al., 2005).

Turbulent motions are represented assuming a Markov process based on the Langevin equation (Thomson, 1987) for each turbulent wind component $\boldsymbol{v}_{t_i}$, $i = 1 \ldots 3$. The formulation includes a drift term and a diffusion term which are functions of the position, the turbulent velocity and time. Cross-correlations between the different turbulent wind components are not taken into account, since they have little effect for long-range dispersion (Uliasz, 1994).

Mesoscale motions (sub-grid motions which are not turbulent in nature) need to be taken into account since they can accelerate the growth of a dispersing plume. Updrafts in convective clouds that occur in conjunction with downdrafts within the clouds and compensating subsidence in the cloud-free surroundings are modelled by a convective parameterization scheme.

2.1.2. Wet deposition

Based on the humidity and temperature from the meteorological input data, the occurrence of clouds is calculated. After that separately in-cloud and below-cloud 'scavenging' of the transported substances are computed in the form of an exponential decay process for the particle mass m:

$$m(t+\Delta t) = m\left(t\right)e^{-\Lambda\Delta t}.$$

For gases the *in-cloud* scavenging coefficient Λ [s^{-1}] is

$$\Lambda = \frac{I}{Hc_{\mathrm{eff}}}$$

where I [mm/h] is the precipitation rate, H is the height over which scavenging takes place and c_{eff} is effective cloud liquid water content.

The *below-cloud* scavenging coefficient is $\Lambda = \mathrm{A}I^{\mathrm{B}}$ where both A and B are empirical parameters specific for each modelled gas. Subgrid variability of the precipitation is also taken into account for the wet deposition via the meteorological data for 'total cloud cover', 'large scale precipitation' and 'convective precipitation' (Stohl et al., 2005).

2.1.3. Dry deposition

The downward flux due to dry deposition F_C of a species with concentration C at height z is described by a deposition velocity.

$$v_d(z) = -F_C/C(z).$$

For gases the deposition velocity is represented as the inverse of the sum of the following 'resistances'

$$|v_d(z)| = [r_a(z) + r_b + r_c]^{-1}$$

where r_a is the aerodynamic resistance between z and the surface, r_b is the quasilaminar sublayer resistance, and r_c is the bulk surface resistance. These are calculated from the atmospheric boundary layer properties contained in the meteorological input data and from the land-use surface data within FLEXPART.

2.2. Required input data

The FLEXPART algorithm needs 3-dimensional, time-dependent meteorological and emissions data as input for the investigated geographical region and time period. The weather data include wind velocities, humidity, temperature, pressure, sunshine, precipitation and turbulence in the atmospheric boundary layer. The emissions inputs are in the form of a list of 4-dimensional space-