

Long-range transport of ammonia released in a major chemical accident at Ionava, Lithuania

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Abstract

On 20 March 1989 a very severe accident occurred at a chemical plant near the town of Ionava in Central Lithuania. We have estimated the atmospheric dispersion of ammonia released in the accident, focussing on the possible long-range effects. The computations were made using trajectory and dispersion models based on gradient-transfer diffusion theory, using actual meteorological data. The computed concentrations were compared with available observations, obtained from the European Monitoring and Evaluation Programme stations.

1. Introduction

In this paper we evaluate the atmospheric dispersion of ammonia released in a major chemical accident near the town of Ionava in central Lithuania. Seven people were killed, 57 injured and about 32 000 evacuated in the accident. Reports on the course of the accident have been prepared by the Lithuanian State Committee for Environmental Protection [1] and by Andersson [2], but these are not published; a summary of this work was published by Kletz [3].

The accident occurred at the chemical plant "Azot", which produces mainly fertilizers. This plant is situated about five kilometres northeast of the town of Ionava. A 10 000 tonne capacity tank containing 7000 tonnes of refrigerated ammonia at its boiling point (-33°C) split without warning. The rupture was caused by an erroneous filling of the tank with relatively warm ($+10^{\circ}\text{C}$) liquid ammonia which formed a layer at the base of the tank. The warm ammonia

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then suddenly rose to the surface and evaporated, and the pressure rise overwhelmed the relief valves. All refrigeration compressors were out of operation [2, 3].

The liquid ammonia released formed a pool, which was in places 70 cm deep. According to the chemical plant itself, about 1400 tonnes of the spilled liquid ammonia evaporated. The pool caught fire, and the fire spread to a fertilizer store containing 15 000 tonnes of NPK 11-11-11. It was theoretically estimated that a further 700 tonnes of ammonia was released into the environment due to the self-sustaining thermal decomposition of this material [1]. Substantial amounts of nitrogen oxides were also released from the fire.

The objective of this study was to analyse whether a release of this magnitude might cause long-range effects, on a scale of tens or hundreds of kilometers. As southerly winds prevailed at the time of the accident, we were interested in determining whether the accident could be observed in Finland, about 500 km north of the accident location.

More generally, the long-range consequences of major accidents should be known in formulating international conventions on transboundary transport of chemically toxic substances. Studying the dispersion of materials released in major accidents may also improve our understanding of the basic atmospheric transport processes, as the research following the Chernobyl accident has demonstrated.

Our computations are based on the release estimates provided by the authorities [1]. Unfortunately the available data was not sufficiently accurate for the application of mathematical models governing liquid pool evaporation and spreading (for instance, [4]). An evaporating liquid pool of ammonia does not produce a heavier-than-air gas cloud, as ammonia vapour at its boiling point is lighter than air at commonly occurring ambient temperatures (0–20 °C). A heavy gas cloud could therefore be formed only if there was significant aerosol formation, which we considered to be unlikely in the reported conditions.

We have therefore applied a long-range dispersion model, which uses a Gaussian plume model up to a specified transition distance, and subsequently the gradient-transfer diffusion theory. Our analyses address mainly downwind distances larger than about 10 km. As the release estimates contain fairly large uncertainties, computations were made for two release scenarios. Our objective has been to obtain order-of-magnitude estimates of the actual concentrations.

The analysis of the meteorological situation is based on radiological sounding data and synoptical observations. Air parcel trajectories and contaminant concentrations were computed using selected release scenarios and numerically analysed meteorological data.

2. Assumed release scenarios

The exact time of occurrence of the accident was 8.15 UTC (Universal Time Coordinated) 20 March 1989 (this time corresponds to 10.15 local Lithuanian

time). Hereafter all times given in this paper are expressed as UTC. Intensive evaporation of the liquid pool lasted for up to about eight hours, and the fire was totally extinguished 17.00 on 22 March [1].

The fire also resulted in releases of nitrogen oxides, fluoride and chlorine into the air, and escape of liquid pollutants into a nearby river. Some measurements of the concentrations in the air for the above-mentioned three substances are available [1]. However, as their released amounts have not been quantitatively estimated in the available references, we address only the dispersion of ammonia in the air in the following. We also neglect the ammonia releases from the fire.

In Scenario 1 we assume that the total amount of ammonia vapour released into the air was 1400 tons and that the duration of the release was eight hours. However, as the rate of ammonia evaporation from the pool most likely decreased with time, most of the ammonia may have been evaporated within a much shorter time. We therefore assume in Scenario 2 a release of 1000 tons of ammonia within one hour. In both cases the release height is assumed to be negligible.

Scenario 1 is a 'best guess' estimate according to available release data [1], while Scenario (2) is a 'worst case' estimate for the release of ammonia into the air from the liquid pool. As the time-dependence of the emission rate is not known, we have assumed it to be constant during the period of release, in both cases.

3. Computational methods

We have applied a long-range dispersion model, which uses gradient-transfer diffusion theory for vertical dispersion. In particular, this approach more readily takes into account the change of the vertical concentration profile brought about by the dry deposition, compared to the Gaussian approach. The model has been described in detail in [5-8]; an overview of the model and its application will be given in the following.

The model applies a Gaussian plume model up to a specified transition distance, which is dependent on atmospheric stability: 200 m for unstable, 500 m for neutral and 1000 m for stable stratification, respectively. Outside this nearby zone the gradient-transfer approach is used for computing the diffusion in the vertical direction.

Assuming steady-state conditions and neglecting diffusion in the downwind direction, we can write the atmospheric diffusion equation in the form [8]

$$U(z) \frac{\partial \bar{C}_z(x, z)}{\partial x} = \frac{\partial}{\partial z} \left[K_z(z) \frac{\partial \bar{C}_z(x, z)}{\partial z} \right], \quad (1)$$

where x is the downwind distance, z is the height, U is the mean wind speed, \bar{C}_z is the relative vertical concentration and K_z is the vertical eddy diffusion

coefficient. The profiles of the vertical eddy diffusion coefficients for different stability categories were estimated using the correlations presented in [9].

The boundary conditions for eq. (1) are as follows

$$\left[K_z(z) \frac{\partial \bar{C}_z(x, z)}{\partial z} \right]_{z=z_0} = v_d \bar{C}_z(x, z_0) \quad (2)$$

and

$$\left[K_z(z) \frac{\partial \bar{C}_z(x, z)}{\partial z} \right]_{z=z_i} = 0, \quad (3)$$

where z_0 is a reference height near ground level, z_i is the mixing height and v_d is the dry deposition velocity. Condition (2) assumes that the vertical mass flux is proportional to the concentration at a reference level near the ground, and condition (3) assumes that the vertical mass flux vanishes at the upper boundary.

Horizontal diffusion outside the nearby zone is also computed using a Gaussian approach with correlations of the dispersion coefficients presented by Hanna et al. [10], up to the distance of 100 km. These correlations were modified to allow for a reduced diffusion rate for longer distances:

$$\sigma_y(x) = Ax^{1/2}, \quad (4)$$

where the factor A was determined by matching $\sigma_y(x)$ with the above mentioned correlations at the distance of 100 km [11]. The model also includes a treatment of wet deposition.

The use of the Gaussian approach for horizontal diffusion has its limitations. In particular, the influence of synoptic scale atmospheric turbulence on the horizontal diffusion cannot be properly taken into account. For the longer release scenario (with a release duration of eight hours) this might be important. However, in the actual meteorological conditions of this study, the divergence between successive trajectories was fairly small, and consequently, the effect of synoptic scale fluctuations is not large.

The atmospheric transformation and deposition processes of ammonia have been addressed in [12–16]. In European conditions most of the gaseous ammonia, NH_3 , will react with H_2SO_4 to form ammonium, NH_4^+ . The conversion rate of this reaction depends on the concentrations of acidic components and on the atmospheric mixing characteristics. A value of 29% per hour has been suggested, based on a comparison of predicted and measured concentrations [14], although somewhat smaller values have also been used [15].

The dry deposition velocity of ammonia vapour depends on atmospheric conditions and the surface cover. Average values from 0.8 to 1.2 cm/s have been used in various models [12–15]. Atmospheric measurements have shown that most ammonium particles are found in fine particles, with diameters from 0.1 to 1.0 μm [14, 17]. An average dry deposition velocity for ammonium of 0.1 cm/s

has been adopted in several models [15]; values of this order of magnitude have also been observed experimentally [16].

Consequently, most of the NH_3 is transformed into NH_4^+ on a time-scale of a few hours, but ammonium may be transported long distances due to its small deposition velocity. Ammonia and NH_4^+ are called NH_x components. These are mainly removed from the atmosphere by dry deposition of NH_3 close to the source, and by wet deposition of NH_4^+ at some distance from the source [12].

The above-mentioned results on the transformation and deposition of ammonia have been obtained for concentrations commonly occurring in the atmosphere, and they may not be directly applicable to the high concentration levels occurring in accident conditions. For instance, the conversion rate of NH_3 to form NH_4^+ may be reduced at high ammonia concentrations.

We have used in the computations a constant deposition velocity appropriate for ammonia vapour, 1.0 cm/s. At long distances this choice overpredicts dry deposition, as part of the NH_x is in the form of particulate ammonium. However, the resulting error is moderate as dry deposition is dominated by ammonia vapour deposition.

Ammonia is very soluble in water, and the occurrence of precipitation may cause a substantial decrease in concentration. However, in the particular meteorological conditions of this study, there was no precipitation during the transport of pollutants to a downwind distance of up to about 500 km.

Air parcel trajectories were computed on a two-dimensional horizontal grid using numerically analysed meteorological data. The trajectory model has been described in detail in [7]. The meteorological data analyses are based on simultaneous synoptical surface observations and sounding and satellite data, from the whole of the northern hemisphere. This data is numerically analysed onto constant pressure levels on a rectangular mesh system. The mesh width is about 150 km, and the values within each grid square are computed by linear interpolation procedures.

By combining trajectories at different levels we can study the transport of air masses in the whole of the atmospheric surface layer. The variation of transport conditions in time and space are also allowed for in these computations. As the pollutants considered here were released near ground level, we have made most of the computations for the 1000 hPa pressure level, corresponding approximately to the lowest 200–300 m layer of the atmosphere. Some computations were also made for the 950 hPa pressure level, corresponding approximately to a height of 500 m.

4. Results

4.1 Meteorological conditions during transport

Dispersion conditions change with time and location during transport. In particular, the time of day and the nature of the underlying surface affect the meteorological situation. For instance, stable atmospheric conditions are more

frequent over the sea, compared to land surfaces. The dispersion conditions therefore vary along each particular trajectory.

We address first the dispersion conditions from the accident location to the southern coast of Finland. The corresponding straight-line distance is about 500 km (Fig. 1). No ice cover existed over the Gulf of Finland (situated between Finland and Estonia) at the time, except for sea areas very near the coastline.

On 20 March 1989, light southerly winds (wind speed at 10 m height ≤ 2 m/s) prevailed in the accident area. In general, during the transport of pollutants neutral and stable atmospheric stability conditions were dominant and mainly moderate southerly winds (2–7 m/s at 10 m height) prevailed. No significant rainfall took place during the transport. Consequently, atmospheric conditions were unfavourable for the rapid mixing of contaminants.

We have selected two characteristic dispersion conditions for our analyses: (i) stable atmospheric stratification (Pasquill-class F) with a wind speed of 2 m/s, and (ii) neutral atmospheric stratification (Pasquill-class D) with a wind speed of 5 m/s. The mixing layer was estimated to be 200 m for stable and 500 m for neutral stratification.

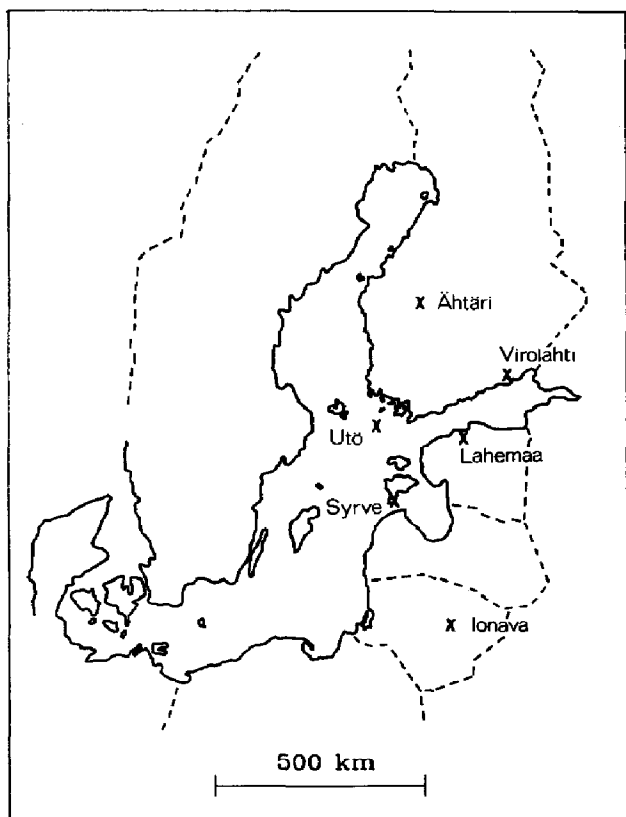


Fig. 1. The accident location and the relevant European Monitoring and Evaluation Programme (EMEP) stations.

Clearly, the actual meteorological conditions vary continuously during the transport of pollutants. For instance, the atmospheric stabilities and the mixing heights depend on the day–night transition, and the structure of the underlying surface. However, most of the time the actual dispersion conditions varied between the two selected cases. Atmospheric dispersion was then analysed as a function of the straight-line downwind distance, for the two release scenarios and the two characteristic dispersion conditions.

We also studied the transport of pollutants over the southern and central parts of Finland. Meteorological considerations show that after the pollutants reached the southern coast of Finland, mechanically and thermally induced atmospheric turbulence and also wind velocity increased substantially. There was also an area of continuous, although weak, rainfall over southern Finland at the time. The pollutant concentrations were therefore reduced much more rapidly after passing the southern Finnish coastline.

4.2 Trajectories of air masses

The source-oriented trajectories of air masses are shown in Figs. 2a–c. The time of occurrence of the accident was 8.15 UTC on 20 March 1989. The trajectories of Fig. 2a cover the period of intensive evaporation of the liquid ammonia pool, while the trajectories of Figs. 2a–c cover the duration of the fire and some days after it. Consecutive trajectories have been shown at three-hourly intervals in Fig. 2a, at six-hourly intervals in Fig. 2b and at twelve-hourly intervals in Fig. 2c. Points along each trajectory have been determined at three-hourly intervals.

Trajectory computations contain errors caused by uncertainties in the meteorological measurements and the numerical procedures, including interpolation. Statistical analyses of the adopted method have shown that the maximum total error is about equal to the difference between two consecutive trajectories having a three hour difference in their starting times.

The results of Fig. 2a show that most of the releases from the accident were transported directly to the southern coast of Finland. The scatter of the different trajectories is quite small, reflecting the slowly changing synoptical meteorological situation at the time. The computed transport times along the various trajectories from the accident location to the southern coast of Finland were about 27–36 hours. For instance, air parcels released from ground level on 20 March at times from 06.00 to 21.00 UTC, would reach the southern coast of Finland at between approximately 15.00 on 21 March and 03.00 on 22 March.

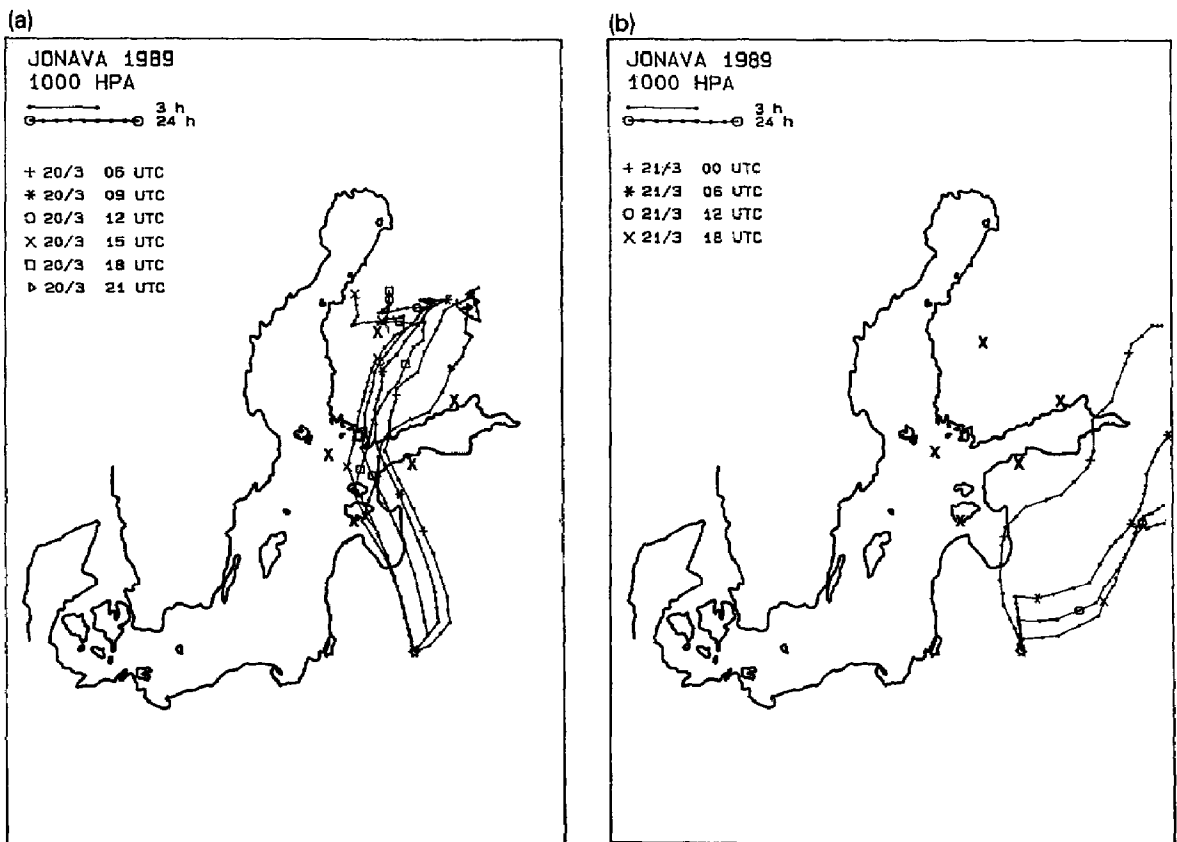
Figures 2b and c show that releases on 21–26 March were directed northeast, east and southeast, into the area of the formerly Soviet Union. The trajectories of Figs. 2a–c were computed for the lowest atmospheric layer, with a height less than about 200–300 m. As some pollutants may have risen to higher altitudes, we also computed the trajectories of air masses in other atmospheric layers. Possible precipitation scavenging from higher layers influences wet deposition measurements.

The results show that the transport directions at higher altitudes turn in a clockwise direction. For instance, air parcels from the accident location on 20 March 1989 are directed at higher layers to the northeast or north to northeast. The contribution of these pollutants to observational data in Finland is therefore fairly small.

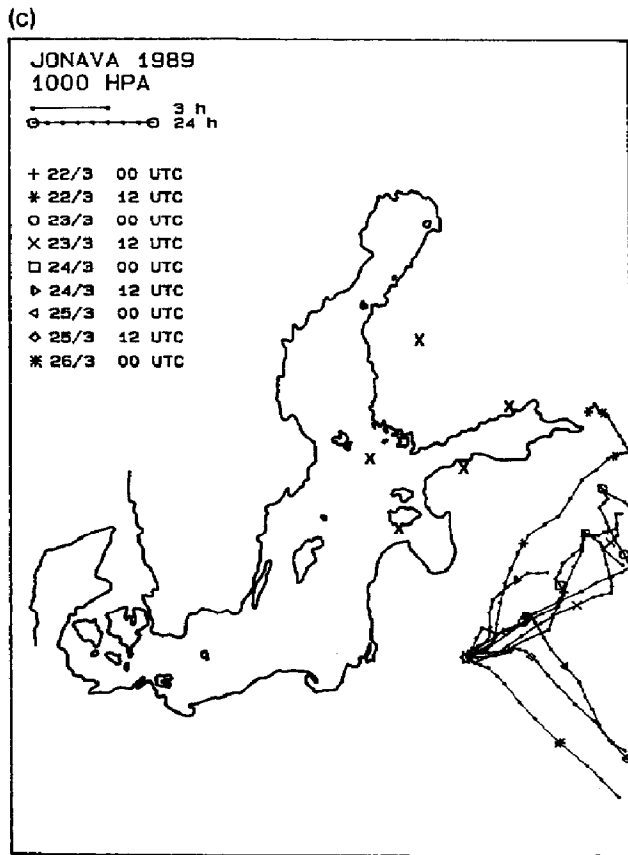
Clearly, the transport times at higher levels are smaller, compared to those at lower levels. Computed transport times from the accident location to the southern coast of Finland along various trajectories at the 925 hPa level were about 13–30 hours. For instance, air parcels released from ground level on 20 March at times between 06.00 and 21.00 would reach the southern coast of Finland at approximately 7.00–12.00 on 21 March.

4.3 Predicted concentrations

Figure 3 shows the maximum concentration on the plume centre-line at the ground level of NH_x components in the air ($\text{NH}_3 + \text{NH}_4^+$) for the selected release scenarios. The order of magnitude of the concentrations at a distance of



Figs. 2a–c. Trajectories in the lowest atmospheric layer (below a height of 200–300 m) originating from the accident site on 20, 21 and 22–26 March 1989. The legend shows the three-hourly and daily intervals and the starting times of the trajectories.



Figs. 2. Continued

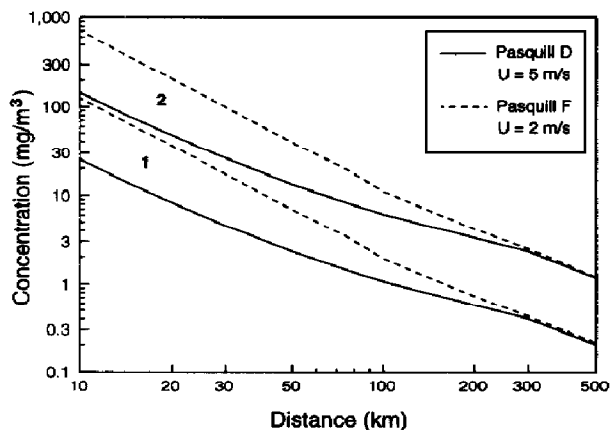


Fig. 3. The contaminant concentration at the ground level as a function of downwind distance for release Scenarios 1 and 2, for two combinations of atmospheric stability and wind velocity.

about 500 km were 0.1–1.0 mg/m³ for Scenario 1 release, and 1–10 mg/m³ for Scenario 2 release.

In these computations we have assumed a constant dry deposition velocity, although it is actually dependent, among other things, on windspeed and atmospheric stability. In particular, the resistance to transfer to the surface for reactive gases increases with decreasing friction velocity, which is typically much lower in stable conditions [18]. Dry deposition velocity is therefore smaller in stable conditions. Dry deposition is also dependent on the roughness length, which depends on the route of the particular trajectory.

A more detailed modelling of the dry deposition process might have an influence on the shape of the curves in Fig. 3. The apparent convergence of the concentration curves for the two stability classes may be fortuitous.

5. Comparison of numerical results and observations

In Finland there are five measurement networks for background concentrations and deposition in Finland, but ammonia measurements are made only in the EMEP stations [19]. EMEP measurements include the concentration of NH_x components in the air and in bulk precipitation samples. Listed from north to south the Finnish stations are: Ähtäri, Virolahti and Utö (Fig. 1). However, NH_x concentrations in the air were not available from the Utö station at the time, and daily measurements were not available from the Estonian stations (Syrve and Lahemaa).

Both the concentration and precipitation samples are collected for 24 hours starting at 6.00. Ammonium and ammonia are collected with oxalic acid impregnated filters (Whatman 40, diameter 47 mm, sampling flow 1 m³/h). The filters are extracted with water, and ammonium, both from the extracts and the precipitation samples, is analysed by the spectrophotometric indophenol method.

Figure 4 and Table 1 show the results of measurements during a period of 30 days in March and April 1989. Both the concentration and wet deposition measurements show a maximum on 21 March 1989 at all stations. The maximum values of wet deposition do not correspond to particularly high values of precipitation. However, a survey of measured values from a longer period and from other years reveals that such maximum values are not uncommon in springtime. The measurement data is published annually; for instance, the reports [19] and [20] present the results in 1989.

For a proper interpretation of these results, one has first to consider the trajectory results. The receptor-oriented trajectories at the Utö station are shown in Figs. 5a–b. These show that a major fraction of the pollutants from the accident did not reach this station. The results also show that the air masses reaching southern Finland at the time originated from Central and Eastern European countries.

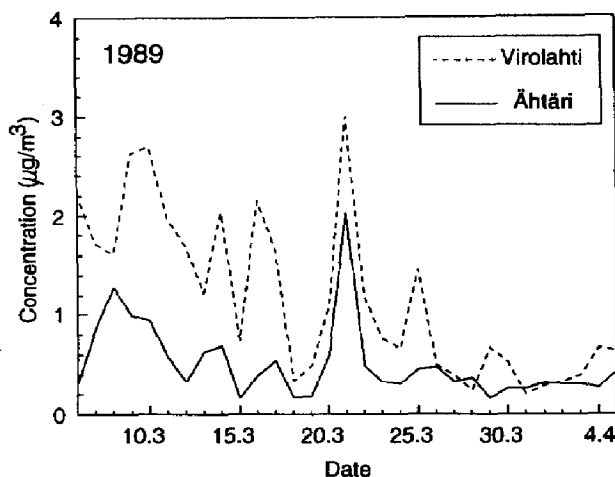


Fig. 4. The measured concentration of ammonia and ammonium in the air as nitrogen ($(\text{NH}_3 + \text{NH}_4^+) - \text{N}$) at two Finnish EMEP stations, two weeks before and after the accident (compiled from [17]).

Similar receptor-oriented trajectories were also computed for the other EMEP stations. Results indicate that some pollutants reached the Ähtäri station. However, as noted earlier, atmospheric dispersion was more efficient over the mainland of Finland, and the contaminant concentrations must therefore have been substantially smaller, compared to concentrations at the southern coast of Finland. Some trajectories reached the Virolahti station, but their contribution does not include the period of efficient pool evaporation.

Detailed trajectory computations showed that in the lowest atmospheric layer (1000 hPa), the times of arrival were about 18.00 on 21 March for Utö, 15.00–18.00 on 22 March for Ähtäri and about 15.00 on 22 March for Virolahti. At the higher transport level (925 hPa) the respective times were 08.00–10.00 on 21 March for Utö, 00.00–08.00 22 March for Ähtäri and 4.00–6.00 on 21 March for Virolahti. Clearly, these figures correspond to trajectories with different starting times and different routes.

Consequently, the trajectory computations indicate that if any detectable amount of NH_x was present it should have been observed in the samples of 21 March at the Utö station, on 22 March at Ähtäri and on 22 March (or 21 March for transport at a higher level) at Virolahti. Maximum values of the concentration and wet deposition measurements were found on 21 March at all stations. The computations are therefore not completely consistent with the measured maximum values.

Our conclusion from the above considerations is that most of the NH_x due to the accident escaped the available measurement stations. The results do not show conclusively that the measured maximum values were caused by the accident.

The Lithuanian authorities reported some ammonia measurements on 20 March 1989 [1]. The largest measured concentration was 200 mg/m^3 about 5 km

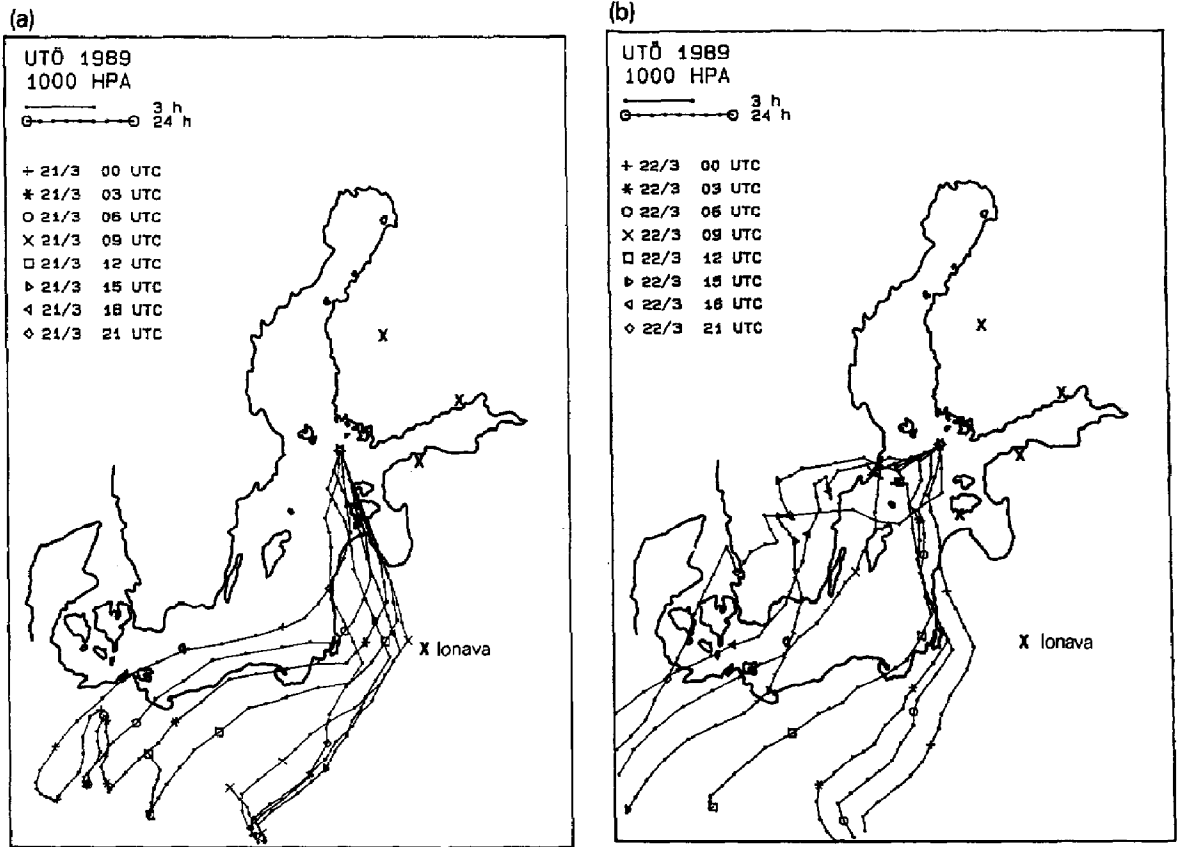
TABLE 1

The measured wet deposition of ammonium, determined as nitrogen ($\text{NH}_4^+ - \text{N}$), and the precipitation at three Finnish EMEP stations, two weeks before and after the accident (compiled from [16]; ND=not detected)

1989 Date	Wet deposition of ammonium (mg N/m^2)			Precipitation (mm)		
	Utö	Virolahti	Ähtäri	Utö	Virolahti	Ähtäri
0603	ND	–	–	0.1	–	–
0703	–	–	–	–	–	–
0803	–	2.4	–	–	1.4	–
0903	–	–	0.1	–	–	0.2
1003	1.5	–	4.4	2.0	–	3.6
1103	14.1	–	3.1	17.0	–	12.9
1203	–	ND	–	–	1.1	–
1303	2.9	–	ND	1.2	–	0.1
1403	ND	3.1	4.8	0.2	2.4	6.5
1503	–	–	–	–	–	–
1603	–	6.7	–	–	2.5	–
1703	ND	–	0.9	0.3	–	1.0
1803	–	–	0.1	–	–	0.7
1903	1.5	1.7	0.6	4.0	2.8	3.4
2003	–	–	0.3	–	–	0.5
2103	6.0	8.4	8.7	5.1	3.8	3.7
2203	2.0	4.4	1.5	3.1	3.9	4.3
2303	1.0	3.2	0.9	2.5	2.5	9.3
2403	1.8	6.6	0.6	4.0	5.0	0.8
2503	ND	3.3	0.9	0.1	2.5	3.7
2603	–	–	–	–	–	–
2703	–	–	–	–	–	–
2803	2.2	5.3	6.1	0.8	2.4	6.2
2903	–	4.5	2.0	–	3.2	5.9
3003	–	–	–	–	–	–
3103	–	–	–	–	–	–
0104	–	–	–	–	–	–
0204	–	–	–	–	–	–
0304	–	–	–	–	–	–
0404	–	–	–	–	–	–
0504	–	–	–	–	–	–

downwind of the accident site at 15.00–16.30 UTC, and concentrations of about 20–25 mg/m^3 were measured at distances of from 5 to 12 km from the source. Further details of the measurement sites and times have not been reported. These values are qualitatively consistent with the concentration curves of release scenario 1 in Fig. 3.

We also received a number of phone calls from individual citizens in Finland after the accident, mainly reporting eye irritation [21]. These observations



Figs. 5a–b. Receptor-oriented trajectories in the lowest atmospheric layer (below a height of 200–300 m) at the EMEP measurement station Utö. The legend shows the three-hourly and daily intervals and the times at which each trajectory reached the station.

were made during the evening of 21 March in a limited area, on or near the southern coastline of Finland, about 10–50 kilometres west of Helsinki. The location and time of these observations are consistent with the results of the trajectory computations in the lowest atmospheric layer (Fig. 2a). We received the phone calls during 22–25 March, before any information about the accident had been disseminated to the public in Finland.

The least detectable odor level of ammonia vapour varies from 1 to 50 ppm, and 20 ppm has caused complaints and discomfort in uninjured workers [22]. It has also been reported that maximum acceptable concentration at the working place without severe complaints is 20–25 ppm [22]. Ammonia vapour causes irritation of eye, skin and mucous membranes. According to the computations, the order of magnitude of the NH_x concentrations on the southern coastline of Finland were 0.1–1.0 mg/m^3 for release Scenario 1, and 1–10 mg/m^3 for release Scenario 2.

We conclude that these observations of irritation may have been caused by the Lithuanian accident.

6. Conclusions

We have analysed the possible long-range effects of an accident at a chemical plant near the town of Ionava, Lithuania, on 20 March 1989. The accident was unusual in several respects. First, the releases into the atmosphere were exceptionally large — the estimated ammonia releases were 1400 tonnes from pool evaporation, and about 700 tonnes from the resulting fire. Intensive evaporation of the liquid pool lasted about eight hours, and the fire continued for three days.

Second, atmospheric conditions were unfavourable for the rapid mixing of contaminants. Neutral or stable atmospheric stability conditions, and mainly moderate or low wind speeds prevailed, and no significant rainfall took place during the transport of pollutants to a distance of up to about 500 km.

The gas cloud was transported directly northwards. Transport times along various trajectories in the lowest atmospheric layer from the accident location to the southern coast of Finland were about 27–36 hours. According to the numerical computations the maximum NH_x concentrations in Finland may have been 0.1–10.0 mg/m^3 . The corresponding commonly occurring background concentrations are in the $\mu\text{g}/\text{m}^3$ range.

Clearly, several uncertainties affect these estimates. The masses of the released pollutants and the release duration are uncertain. The transformation and dry deposition processes were not modelled in detail. The dispersion computations do not include the changes of meteorological conditions with time and location along each trajectory.

The results indicate that most of the NH_x due to the accident escaped the EMEP monitoring stations in Finland. The EMEP network has been designed for monitoring background air quality, and is not well-suited for detecting chemical emergencies. The daily sampling time is too long for such rapidly occurring releases. Only a few of the hundreds of toxic gases commonly used by industry are measured in the EMEP network. A more closely spaced measurement network, equipped with instruments sampling with a sufficient time resolution, would be needed for reliable detection of accidentally released chemical compounds.

Toxic substances released accidentally from industrial processes may be transported substantial distances in the atmosphere. Information that is as accurate and reliable as possible should be disseminated to the public, and over a much wider area than the zone of immediate health effects. International conventions should therefore guarantee, in particular, open and undelayed information exchange in case of such emergencies.

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