of two comparatively large figures, whereas they become very reliable further inland. A similar caution should be exercised with most of the other excess data over or near the ocean. For this reason, we did not use the pattern of the excess concentration maps of Na^+ and K^+ for our considerations. Our line of reasoning was based on the important fact that the average concentrations of Na^+ and K^+ over most of the U.S. are very much the same despite the fact that the ratio K^+/Na^+ is only 0.041 in sea water and thus 20 times smaller than in rain. There is no basis for the assumption that the ionic composition in sea spray particles deviates that much from sea water. On the contrary, coastal rains in unpolluted areas have ratios close to that of sea water. We concluded, therefore, that most of the K^+ must come from the soil. Because of the similarity in chemical behavior of Na^+ and K^+, we suggested that it is very likely that the excess Na^+ is also derived from the soil.

As we pointed out earlier, KALLE (1953—1954) in a very interesting study, provided considerable evidence of the mineral origin of the excess Na^+. K^+, Ca^{++} and other cations in rain over Europe, It would have been very useful if this paper had been included in Eriksson's discussion. Also, the reader would have appreciated a reference of METHAM's (1950) paper on the SO_2 concentration and SO_4^- deposition over England in connection with Figures 7.7 to 7.10. Metham's work indicates that industry is an important, if not the predominant source of sulfur over England, in contrast to the conclusions drawn from Figures 7.7. to 7.10. The heavy rainfalls on the west coast of Ireland certainly increase the deposit, but, as with our Sr^{90} example, this does not indicate that the ocean is the source of sulfur.

Sincerely yours,

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Reply

Dear Sir,

The main question raised by Dr. Junge is no doubt of fundamental importance for the correct interpretation of chemistry data on precipitation, though his analysis perhaps can be extended somewhat. Firstly, as most precipitation comes from disturbances which are moving comparatively slowly we can regard precipitation as a quasi-stationary state where horizontal convergence of moisture is steadily replacing the moisture precipitated. Neglecting contribution to the precipitation by sub-cloud air is probably permissible. Provided also that most of the precipitation is due to accretion of cloud droplets into larger aggregates, i.e. condensation on ice nuclei is quantitatively unimportant, the equation given by Dr. Junge is, of course, correct. But K in his formula is an instantaneous value so it has to be averaged and the way it is done by Dr. Junge may be too simple.

Denoting by m the amount of a substance precipitated, by R the rate of precipitation, and using instead of $1/l$ a specific volume v (volume per gram liquid water) we can write

$$m = \int_0^T \varepsilon R v c \, dt \qquad (2)$$

from which also the average weighted concentration can be derived. The integration is, of course, carried out only during precipitation.

Now we can assume ε to be constant, a reasonable assumption for most compounds of interest. In order to carry out the integration we use a common method writing $R = \bar{R} + R'$, $v = \bar{v} + v'$ and $c = \bar{c} + c'$.

Now an average of the sum of products like $R'v'$ can be written $r\sigma_R\sigma_v \cdot T$ where r is the correlation coefficient between R and v and σ_R and σ_v are standard deviations of R and v respectively. Then we get for the integral

$$m = \varepsilon\left[1 + r_{Rv}\cdot\frac{\sigma_R}{\overline{R}}\cdot\frac{\sigma_v}{\overline{v}} + r_{Rc}\cdot\frac{\sigma_R}{\overline{R}}\cdot\frac{\sigma_c}{\overline{c}} + \right.$$

$$\left. + r_{vc}\frac{\sigma_v}{\overline{v}}\cdot\frac{\sigma_c}{\overline{c}} + r_{Rvc}\cdot\frac{\sigma_R}{\overline{R}}\cdot\frac{\sigma_v}{\overline{v}}\cdot\frac{\sigma_c}{\overline{c}}\right]\cdot\overline{R}\cdot\overline{v}\cdot\overline{c}\cdot T \quad (3)$$

This looks complicated but, including the parenthesis into the constant we get

$$m = \varepsilon'\,\overline{R}\cdot\overline{v}\cdot\overline{c}\cdot T \quad (4)$$

Now ε' depends on the correlations between and the relative variations of R, v and c. It may be constant within a smaller geographical unit but we do not know for certain. It may be different for maritime compared to continental environments. We may assume it to be constant, however, but it may be less than ε or larger than ε depending on whether correlations are negative or positive.

From eq. (4) we then get the average weighted concentration $\overline{c}_p$ in precipitation

$$\overline{c}_p = \varepsilon'\cdot\overline{v}\cdot\overline{c} \quad (5)$$

and if $\overline{v}$ is constant in space $\overline{c}_p$ will be entirely dependent on $\overline{c}$. So when interpreting his maps of concentrations in precipitation Dr. Junge assumes both $\overline{v}$ and ε' to be essentially constant over the whole area. Or, better, their variation does not obscure the linear relation between $\overline{c}_p$ and c too much.

If $\overline{R}$ should be inversely related to $\overline{v}$ which means that the precipitation rate is proportional to the liquid water content in clouds, $\overline{R}\cdot\overline{v}$ should be essentially constant in a given area. As most precipitation comes from large scale disturbances, T over a longer time would also be constant. In addition, if ε' can be regarded as constant m should be proportional to $\overline{c}$. This is then, in effect, the assumption I have made in my paper when interpreting total deposition data. If this assumption is correct, Dr. Junge's assumption must be wrong and vice versa.

Now, the rate of precipitation in large scale disturbances depends on the degree of horizontal convergence which can be amplified through mountains and ground friction. It depends also on the convective processes going on in the clouds but, as these depend upon the release of latent heat through condensation they must depend on the liquid water content. A lower value of v thus increases the convective activity and it is well known that this also causes an entrainment of upper, drier air into the clouds. The average concentration of sea salt is therefore more or less inversely proportional to the depth of the clouds and the same will be true more or less for any element originating at the earth's surface which does not spend too long time in the atmosphere. Even for elements with a relatively long residence time, like sulphur, it will still hold close to the source areas. Hence, for elements originating at the earth's surface one can expect R to be negatively correlated to c and possibly also to v. Then m will be proportional not to $\overline{c}$ but to the total amount per unit area of the element. The concentration in precipitation will, however, be inversely related to the rate of precipitation.

If, however, a chemical element should originate at the tropopause as in the case of Sr^{90} one would expect something entirely different, namely that R and c should be positively correlated, especially as we know that the concentration of Sr^{90} increases with altitude. The effect of this positive correlation may be partly offset by a negative correlation between R and v. In this case $\overline{v}\cdot\overline{c}$ tends to be constant and m well largely depend on the amount of precipitation. This is an interesting point and it is rather significant that at least in Scandinavia the pattern of precipitated radioactive debris does not follow that of precipitated excess sulphur. In fact, nitrate-nitrogen is the only compound that bears any kind of resemblance to Sr^{90} in Scandinavia, suggesting, at least partly, a stratospheric origin.

It is thus seen that the interpretation of chemical data on precipitation is a rather complex problem and, until more direct evidence is available it is a matter of intuition and, perhaps, taste in which way they are interpreted.

Dr. Junge also believes that the excess Na and K computed from this data for places close to the coasts are unreliable because they are obtained as differences between large numbers. However, as they are of the same sign, they cannot be due to random errors in the analyses. There are consequently only two alternatives left, one is that they are real and the other that they are due to systematic errors in the analyses.

Dr. Junge also states that there is no basis for the assumption that the ionic composition of sea spray

particles deviates much from that of sea water. I have discussed some possibilities in my paper and I have the opinion that there is no basis for excluding the assumption that the ionic composition of sea spray particles deviates much from that of sea water.

Finally Dr. Junge brings forth two references which he considers should have been included, and he is correct in that respect. Kalle, whom he refers to first used the data from the first Scandinavian network published by EMANUELSSON, ERIKSSON & EGNÉR in 1954. He too computed the excess Na, K and Ca in the precipitation at the different stations as I have done and plotted them as percentage of the total excess on the familiar triangular diagram together with the average composition of various rocks with respect to these three components.

He finds that the excess Na, K and Ca in precipitation falls outside the region of composition of various rocks. Now, this can be expected as any terrestrial matter must originate at the surface of soils and soils differ significantly in their composition from rocks. Kalle is also aware of this and makes some corrections on the excess to bring it in line with the composition of rocks. This is, of course, useless as rocks in general contribute nothing to the atmosphere, only the soils which are covering the rocks. Now, it seems also evident that the more soluble parts of the soil, at least in the areas which contribute to the atmosphere are dominated by the chemical composition of precipitation but not in a simple way.

• It is interesting though distressing to note Kalle's opinion on the question of the origin of the excess components in precipitation. He states: "Vieles scheint dafür zu sprechen, dass der bisher unerklärliche Vorgang der Jonentrennung bei der Niederschlagsbildung in der Atmosphäre durch den Vermischungsvorgang ozeanischer und litosphärischer Salzanteile über den Kontinenten vorgetäuscht ist und dass es nicht nötig ist, zu völlig neuartigen, zum Teil reichlich hypothetischen Erklärungsversuchen seine Zuflucht nehmen zu müssen." Principally, it is fair to disbelieve new theories and hypothesis, it is even necessary in science but one should never state categorically without documented arguments that there is no need for such theories and hypothesis.

As for Meetham's work I do not share Dr. Junge's belief that industry is the dominant source of sulphur over England. I have also in my paper given some thought to this question. Both Dr. Junge and I agree that the residence time of sulphur in the atmosphere is appreciable, perhaps 2—3 weeks. Now, everybody acquainted to the British climate will have difficulties comprehending how the English industrially released sulphur can be kept within the boundaries of England for that long.

Meetham also computes a residence time of the industrial sulphur with respect to absorption by the ground and finds that it is somewhere between 5 and 16 hours. If it were that short then one could disregard industrial sulphur as contributing to the average concentration of sulphur in the atmosphere. However, apart from these points my view is that Meetham's work is a significant contribution but his interpretation of the data may need some revision.

Sincerely yours,

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REFERENCE:

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"*Some Statistical Aspects of the Dynamical Processes of Growth and Occlusion in Simple Baroclinic Models*", by Philip D. Thompson, *Rossby Memorial Volume*, pp. 350—358, The Rockefeller Institute Press, 1959.

Dear Sir:

In a recent paper by P. D. THOMPSON (1959) appearing in the Rossby Memorial Volume, a statistical dynamical approach was used to investigate the "growth" and "occlusion" in the 2-level baroclinic model. The main conclusions from the investigation are that

1. ' The simple 2-level model has a selective preference for disturbances in which the temperature pattern lags behind the pressure pattern; its dynamical properties are such that disturbances in which the temperature field precedes the pressure field die out and remain undetected, while disturbances in which the temperature field lags behind are amplified."

2. "... through the very same mechanism by which 'out-of-phase' disturbances grow, the temperature field is gradually brought into phase with the pressure field."

Some reservation is apparently taken to the last conclusion in a note at the end of the paper, in which it is pointed out that there exists a stable stationary phase lag between the temperature and pressure fields provided the wave number exceeds a certain critical value.

The conclusions quoted above from Thompson's paper are based on a theory in which the velocity field is independent of the north-south direction and, in addition, that the amplitudes of the disturbances are small. Due to these assumptions one would expect to find an agreement between Thompson's conclusions and the complete solution of the equations for the 2-level model, which can be solved in this particular case as demonstrated by OGURA (1957).

From Ogura's solutions it is first of all seen that there is no "growth" and no "occlusion" in the stable case. The amplitudes will have characteristic periods of fluctuation depending on the vertical windshear and the wave-number as seen from Table 1 in Ogura's paper. The phase-angle will vary monotonically in the stable case as seen from his figure 2.

In the unstable case Ogura has only illustrated one case, in which the disturbance at the upper level initially lags a quarter of a wave-length behind the disturbance at the lower level. It is found that the amplitude increases with time at both levels (fig. 3), and that the phase-angle approaches a limiting phase-angle depending on the vertical windshear and the wave-number (fig. 2). A closer inspection of the solution shows that the phase-angle *independent* of the initial state quite rapidly will approach the limiting phase-angle, which always is such that the disturbance at the upper level is lagging behind the disturbance at the lower level, or in Thompson's formulation: the temperature field is lagging behind the pressure field. With regard to the amplitudes of the disturbances it is found that they may decrease initially if the temperature field precedes the pressure fields, but that amplification sets in when the phase-angle has changed in such a way that the temperature field is lagging behind the pressure field.

The 2-level model has therefore no selective preference for disturbances in which the temperature pattern lags behind the pressure pattern in the sense that disturbances with the opposite phase lag will die out. It is rather a result of the development of the flow pattern in the unstable case that the disturbances will have the temperature field lagging behind the pressure field. Another indication that the 2-level model has no selective preference is the fact that the instability criterion for this model only depends on the wave-number, the vertical windshear, the static stability and the Coriolis parameter. It has thus no reference to the initial arrangement of the temperature field relative to the pressure field.

Concluding the comparison between Ogura's and Thompson's conclusions it is evident that the development of the flow in predictions with the 2-level model, at least in the case where the flow is independent of the north-south direction, does not approach a state of quasi-barotropy, and that Thompson's equations for "growth" and "occlusion" at best can give only the initial changes of the amplitude and phase-angle of the disturbance. Thompson's conclusions are further in disagreement with recent calculations of the conversion between potential and kinetic energy based on the 2-level model as performed by SALTZMAN (1959) and the present writer (1959), in which it is shown that the conversion at any time is from potential to kinetic energy. This result can only be explained if the temperature field systematically lags behind the pressure field.

Very truly yours,

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Reply

Dear Sir:

Several points of criticism implied (but charitably unstated) in Dr. Wiin-Nielsen's letter I find to be very well taken. I have been uneasy about these points for some time and welcome this opportunity to set them straight.

The conclusions quoted by Dr. Wiin-Nielsen, taken literally and as they stand, are correct only to the extent that they correctly relate the rate of growth to the current phase-lag; the error lay in misinterpreting my own equations and, in particular, in supposing that the stationary states of amplitude and phase-lag are *asymptotic* states, in the sense that the system inevitably tends toward those states. In fact, I could not have resolved the latter question, simply because my formulation of the problem was then incomplete.

A mathematically complete description of the behavior of the linear two-level model in question (sinusoidal perturbations of small amplitude, super-